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Application of Taguchi Method and Orthogonal Arrays for Optimization of Adhesion of $SrZrO_3$ Coatings on Ag/Bi(2223) Tapes

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***Abstract*--** **Adhesion of $SrZrO_3$ resistive oxide barrier on Ag sheathed Bi(2223) tapes prepared by the sol-gel and dip-coating method was evaluated with an aid of Taguchi method and $L_{18}(2^1\times3^7)$ orthogonal arrays to determine the optimal process combination of levels of factors that best satisfy the bigger is better quality characteristic (QC=B). For analyses of results, statistical calculations such as average and analysis of variance (ANOVA) were employed to analyze the results for improving the performance qualities of the dip-coated $SrZrO_3$ film. Experimentally, the performance of the films was evaluated in terms of bond strength by varying Sr/Zr mol ratio (A), amount of organic vehicle additives (B), drying temperature (C) and time (D), heat treatment temperature (E) and time (F), respectively. The optimal combination of levels of factors was determined to be $A_3B_2C_3D_2E_1F_3$ having a 90% confidence level.**

1. INTRODUCTION

Bi(2223) superconducting multifilamentary tapes prepared by powder-in-tube process suffer alternative current (ac) losses caused by the magnetic interaction between two neighboring parallel superconducting tapes carrying a transport current because they are exposed to alternative currents (ac) and fields [1],[2]. The use of the Ag sheath material enhances the considerable coupling currents flowing in the sheath under ac magnetic fields [2]. The solutions to reduce ac losses are to subdivide the superconducting materials into many small filaments and the introduction of resistive ceramic oxide barriers between the filaments [3]. However, the effect of the former process is largely cancelled by currents which are induced in the low-resistive Ag matrix. Therefore, the formation of highly resistive oxide barriers around the Ag-sheathed Bi(2223) tapes has been considered as the method of choice for ac applications such as transformers, motors, and power transmission lines.

The ceramic oxide barrier should be an insulator, stable under Bi(2223) annealing conditions and easy to deform. Also, the oxide layer should have a uniform thickness and a good adhesion with the Ag sheathed superconducting tape to prevent the sausage effect during the multifilament fabrication such as drawing and rolling and to maintain the overall critical current density. Witz et al. [2] reported that $CaZrO_3$ and $PbZrO_3$ reacted with Bi,Pb(2223) during heat treatment, resulting in an excess of Ca or Pb and a deficiency of Sr inside the filaments. And similar behavior was observed for $CaWO_4$, $PbWO_4$, $CaMoO_4$ and $PbMoO_4$. They also argued that $SrTiO_3$ and commercial $BaZrO_3$ reduced the Bi,Pb(2223) formation rate, suggesting that those oxides were not adequate for the barrier. On the other hand, $SrZrO_3$ was proposed as a prospective oxide barrier because it had no influence on the Bi,Pb(2223) formation rate and no chemical reaction with the filaments [2]. $SrZrO_3$, which is commercially available and cheap, has been known to have a high mechanical and chemical stability [4].

$SrZrO_3$ resistive oxide barriers on Ag sheathed Bi(2223) tapes were prepared by the sol-gel and dip coating method. Organic vehicle (ethyl cellulose) was added to the $SrZrO_3$ sol-gel to enhance adherence (bond strength). The quality engineering method developed by Dr. Taguchi has been applied to determine the optimal combination of levels of factors that best satisfy the bigger is better quality characteristic (QC=B). To meet the purpose of determining the design solutions, the use of signal-to-noise ratio (S/N) for analysis of repeated results, which is the mathematical formula used to calculate the design robustness, may give a sense of how close the design is to the optimum performance of a process. Experiments with 6 factors at 3 levels were accomplished using the $L_{18}(2^1\times3^7)$ orthogonal arrays to lay out experiments of particular factor constituents. For analysis of results, statistical calculation such as average and

analysis of variance (ANOVA) were employed. In the present study, 6 three-level factors were considered as follows: (A) Sr/Zr mol ratio; (B) amount of ethyl cellulose; (C) drying temperature; (D) drying time; (E) heat treatment temperature; and (F) heat treatment time.

2. EXPERIMENTAL PROCEDURES

Ag sheathed Bi(2223) tape having a dimension of 20㎜ ×3㎜ was polished using a SiC grit of 600 and then surface treated using ultrasonic cleaner using distilled water. The $SrZrO_3$ precursor solutions were prepared from commercial reagent grade strontium acetate-hemihydrate ($Sr(CH_3COO)_2\cdot 1/2H_2O$, Junsei, Japan) in glacial acetic acid by stirring and heating to 80°C. Zirconium(IV) propoxide (~70% in propanol, Fluka, Japan) was mixed with glacial acetic acid and then acetylacetone (99%, Junsei, Japan) was added [5]. Both solutions were mixed and stirred at 80°C and then the resulting solution was diluted with distilled water to form 0.5 M of $SrZrO_3$ precursor solution. Organic vehicle (ethyl cellulose, 100 cps, Kan, Japan) dissolved in α-terpineol (>95%, Kan, Japan), which was prepared at 60°C, was added to the $SrZrO_3$ precursor solution by weight. The solutions were mixed and stirred for 1 h at room temperature.

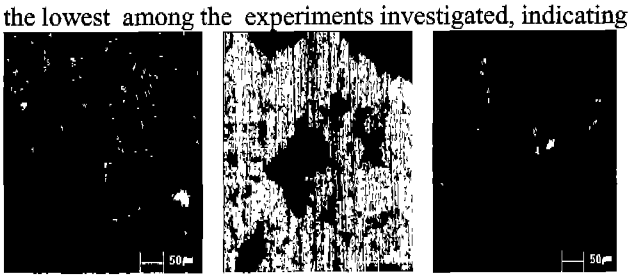
Thin films were obtained from the precursor solution by successive dip coating on Ag sheathed Bi(2223) tape. The films were dried for 5 ~ 10 min at 100 ~ 130°C and then heat treated for 10 ~ 20 min at 500 ~ 700°C to remove organic residues such as alkoxy groups and to form chemical bonds between $SrZrO_3$ coating layer and Ag sheathed Bi(2223) tape [5]. This process was repeated successively up to 4 times.

Microstructure and phase of the film were observed by scanning electron microscopy (SEM, S-2400, Hitachi, Japan) and XRD (3000PTS, Seifert, Germany), respectively. Bond strength of the films was evaluated by a tape test according to ASTM D3359-95a [6].

3. RESULTS AND DISCUSSION

Bond strength of $SrZrO_3$ films was investigated by a tape test according to ASTM D3359-95a to evaluate the effect of organic vehicle addition on adherence between the films and the Ag sheathed Bi(2223) tapes. The extent of adherence between the films and the tapes was analyzed by using a computer image mapping. The white area as shown in Fig. 1 indicated the portion of the detached area after the tape test. Experimental bond strength data were converted to S/N ratio having the properties of QC=B according to the equation of $S/N=-10\times\log[1/n\Sigma(1/j^2)]$, where n and j are the degree of freedom and the bond strength, respectively, as listed in Table I. Factor and level descriptions are summarized in Table II. 6 three-level factors and 2 dummy factors were used in the present study. S/N values having the same factor and level were summed up to examine the ranks of variables and analyze the ranks instead of the original values [5],[7],[8]. The optimal experimental condition can be achieved when the S/N ratio becomes the largest among the experiments investigated. The optimal combination of levels of factors were $A_3B_2C_1D_3E_1F_2$ (No. 17) and $A_3B_2C_3D_2E_1F_3$ (No. 8). The average and the contribution rate of individual levels were calculated based on the S/N ratio, which is shown in Table III. The variation of the average values of levels increased as the experimental factors were varied from F→B→C→D→E→A, indicating that the influence of experimental factors on the bond strength became pronounced in the order of F(1.00)<B(4.44)<C(4.50)<D(4.91)<E(5.17)<A(22.86). It suggested that the subdivided levels were directly related to the bond strength of the $SrZrO_3$ film, therefore, small deviation of the contribution rate (factor A, Sr/Zr mol ratio) having a higher value in Table III was more susceptible to the large divergence of the bond strength.

Square sums of S/N values divided by the number of experiments having the same factor and level (S), degree of freedom (φ) and mean squares (V=S/φ), are summarized in Table IV. In Table IV, one of V values was incorporated into error term (pooling) because V value of factor F was the lowest among the experiments investigated, indicating



(a) (b) (c)

Fig. 1. Surface photographs of the $SrZrO_3$ film of (a) No. 8, (b) No. 11, and (b) No. 17 specimens after tape test.

that the contribution of F to the bond strength of the $SrZrO_3$ film may be insignificant. ANOVA analysis results after pooling are listed in Table V. Ratio of variance (or F-ratio, $V_{factor}/V_e=F_o$) was calculated and then compared with the theoretical $F(\varphi_{factor},\varphi_{error};\alpha=0.1)$ value determined from the standard table in the ref. 9. The lowest F_o value of 9.59 (factor C) in the present study was examined in order to evaluate the reliability of the coating process. F_o value of factor C was greater than that (9.29) of $F(2,5;\alpha=0.1)$, implying that the influence for the factors in Table 5 was significant with a 90% confidence level.

The top-coat film was analyzed using XRD to identify $SrZrO_3$ phase as shown in Fig. 2. Although the S/N ratio of No. 17 in Table I was the highest, no $SrZrO_3$ phase was observed, indicating that $SrZrO_3$ of the No. 17 specimen was not crystallized. Therefore, No. 8 specimen having the second highest S/N ratio was likely to be the optimal combination of levels of factor ($A_3B_2C_3D_2E_1F_3$) for the coatings. Thus, the dip-coated $SrZrO_3$ film having 0.3/0.7 of Sr/Zr mol ratio and 5 wt% of ethyl cellulose, dried for 10 min at 160°C, and subsequently heat treated for 20 min at 500°C, was likely to be the optimal experimental condition.

Fig. 3 is a SEM image of the surface of the $SrZrO_3$ coating (No. 8 and 17), which demonstrates that the crack-free $SrZrO_3$ is composed of small grains with diameters in the range of 0.7 μm to 5 μm. SEM micrographs of cross sections of the $SrZrO_3$ coating layers are depicted in Fig. 4. Although it was difficult to obtain clear and sharp images due to the sample preparation processes (cutting and polishing), a clear interface between the $SrZrO_3$ coating and the Ag sheath was observed, indicating that no chemical interaction between the $SrZrO_3$ coating and Bi(2223) occurred. The grain growth occurred for the specimen of No. 8 ($A_3B_2C_3D_2E_1F_3$), however, the white small grains were still visible for No. 17 specimen ($A_3B_2C_1D_3E_1F_2$), indicating that the crystallization of $SrZrO_3$ was not completed as demonstrated by XRD results. The influence of experimental factors on the bond strength was in the order of $F<B<C<D<E<A$. Therefore, it is conceivable that drying time and temperature (factors D and C) may govern the crystallization of $SrZrO_3$ because both specimens were prepared under the same condition of A, B and E factors and the contribution of F to bond strength of the $SrZrO_3$ film may be insignificant.

TABLE I
EXPERIMENTAL DESCRIPTIONS USING LEVEL NOTATIONS FOR $L_{18}(2^1X3^7)$ AND THEIR S/N VALUES

No.	e	A	B	C	D	E	F	e	S/N
1	-	1	1	1	1	1	1	-	14.72
2	-	1	2	2	2	2	2	-	14.78
3	-	1	3	3	3	3	3	-	14.32
4	-	2	1	1	2	2	3	-	16.39
5	-	2	2	2	3	3	1	-	15.31
6	-	2	3	3	1	1	2	-	14.37
7	-	3	1	2	1	3	2	-	20.92
8	-	3	2	3	2	1	3	-	28.97
9	-	3	3	1	3	2	1	-	26.58
10	-	1	1	3	3	2	2	-	14.11
11	-	1	2	1	1	3	3	-	12.65
12	-	1	3	2	2	1	1	-	15.39
13	-	2	1	2	3	1	3	-	14.11
14	-	2	2	3	1	2	1	-	15.46
15	-	2	3	1	2	3	2	-	15.26
16	-	3	1	3	2	3	1	-	24.56
17	-	3	2	1	3	1	2	-	30.97
18	-	3	3	2	1	2	3	-	22.57
Total sums									331.44

TABLE II
FACTOR AND LEVEL DESCRIPTIONS

Level	e	A	B	C	D	E	F	e
1	-	0.7/0.3	3	100	5	500	10	-
2	-	0.5/0.5	5	130	10	600	15	-
3		0.3/0.7	7	160	15	700	20	-

TABLE III
AVERAGE AND CONTRIBUTION RATE OF INDIVIDUAL LEVELS

Individual level	Average	Contribution rate
A_1	28.66	10.25
A_2	30.30	11.89
A_3	51.52	33.11
B_1	34.94	16.53
B_2	39.38	20.97
B_3	36.16	17.75
C_1	38.86	20.45
C_2	34.36	15.95
C_3	37.26	18.85
D_1	33.56	15.15
D_2	38.45	20.04
D_3	38.47	20.06
E_1	39.51	21.10
E_2	36.63	18.22
E_3	34.34	15.93
F_1	37.34	18.93
F_2	36.80	18.39
F_3	36.34	17.93

TABLE IV
ANOVA ANALYSIS BEFORE POOLING

Factor	S	φ	V
e	0.09	1	0.09
A	488	2	2.44
B	16.87	2	8.44
C	16.50	2	8.25
D	23.96	2	11.98
E	20.13	2	10.07
F	0.76	2	0.38
e	3.46	2	1.73
total	569.77	15	284.94

TABLE V
ANOVA ANALYSIS BEFORE POOLING

Factor	S	φ	V	F_o	F(0.1)
A	488	2	244	283.72	9.29
B	16.87	2	8.44	9.81	9.29
C	16.50	2	8.25	9.59	9.29
D	23.96	2	11.98	13.9	9.29
E	20.13	2	10.07	11.7	9.29
error	4.31	5	0.86	-	
total	569.77	15	284.94	-	

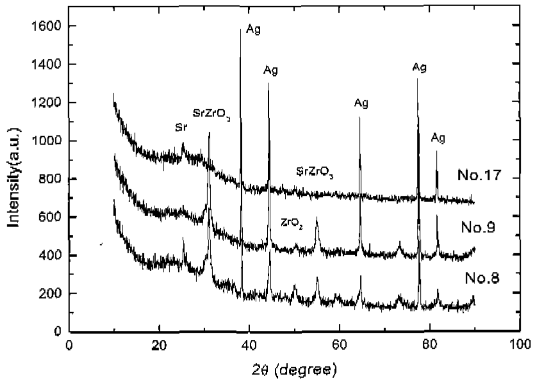
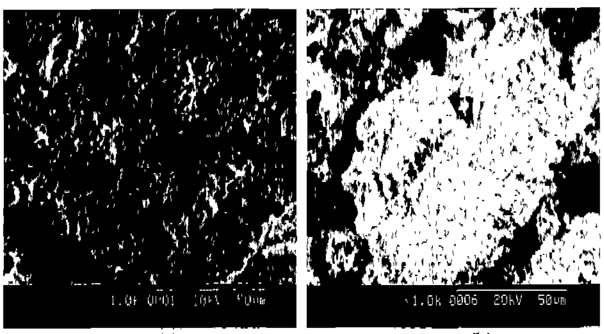


Fig. 2. XRD patterns of the $SrZrO_3$ thin films on Ag sheathed Bi(2223) tapes.



(a) (b)

Fig. 3. SEM micrographs of surfaces of the $SrZrO_3$ thin films: (a) No. 8 and (b) No. 17 specimens, respectively.

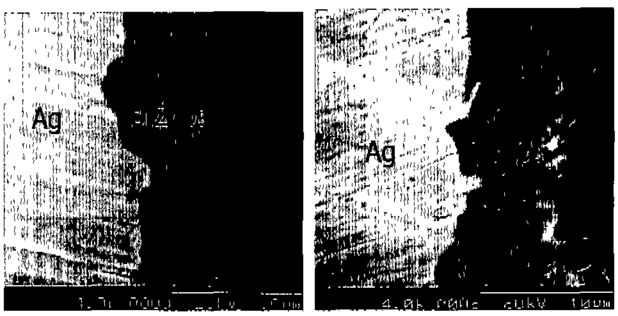


Fig. 4. SEM micrographs of cross sections of the $SrZrO_3$ thin films: (a) No. 8 and (b) No. 17 specimens, respectively

4. Conclusions

The optimum process condition of the $SrZrO_3$ film on Ag/Bi(2223) films for the bond strength was evaluated by Taguchi method and orthogonal arrays and determined. Experimentally, the dip-coated $SrZrO_3$ film ($A_3B_2C_3D_2E_1F_3$) was composed of 0.3/0.7 Sr/Zr mol ratio, 5 wt% of ethyl cellulose, drying temperature and time of 160°C and 10 min, heat treatment temperature and time of 500°C and 20 min, respectively. The variation of the average values of levels decreased as the experimental factors were varied from A→E→D→C→B→F, indicating that the influence of Sr/Zr mol ratio(A) on the bond strength became the most pronounced. In the present study, the influence for the factors was significant with a 90% confidence level and the control parameter of $SrZrO_3$ crystallization was determined to be drying time and temperature (factors D and C).

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