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RESIDUAL STRESS ANALYSIS OF INSULATION COATINGS FOR MAGNET TECHNOLOGIES

ANALIZA NAPRĘŻEŃ SZCZĄTKOWYCH W IZOLACYJNYCH POWŁOKACH OCHRONNYCH DO ZASTOSOWAŃ W MAGNESACH

High temperature $MgO\text{-}ZrO_2$ insulation coatings were grown on long-length Stainless-Steel (SS) tapes by reel-to-reel sol-gel method for applications of High Temperature Superconductor/Low Temperature Superconductor (HTS/LTS) coils and magnets. The residual stresses were investigated analytically at various temperature ranges, 580°C to 25°C (room temperature) and -196°C (liquid helium temperature), and 630°C to 25°C and -196°C for different thicknesses to be 13, 12, and 7 μm. The maximum stress values were obtained to be 1.92 GPa in tension for SS substrate with the 13 μm coating and -2.42 GPa in compression for the 7 μm $MgO\text{-}ZrO_2$ coating in the temperature range between 630°C and liquid helium temperature. The surface morphologies and microstructure of sample were also characterized using a scanning electron microscope (SEM). SEM observations revealed that $MgO\text{-}ZrO_2$ coatings have a mosaic like crack structure.

Keywords: residual stress, sol-gel, $MgO\text{-}ZrO_2$, insulation and superconducting magnet

Wysokoemperaturowe powłoki izolacyjne $MgO\text{-}ZrO_2$ wytworzono na długich taśmach ze stali nierdzewnej metodą reel-to-reel zol-żel dla zastosowań w cewkach i magnesach typu wysokotemperaturowy nadprzewodnik/niskotemperaturowy nadprzewodnik. Naprężenia badano analitycznie w różnych zakresach temperatur, 580°C do 25°C (temperatura pokojowa) i -196°C (temperatura ciekłego helu), oraz 630°C do 25°C i -196°C dla różnych grubości powłoki: 13, 12 i 7 μm. Maksymalne wartości naprężeń otrzymano w zakresie temperatur pomiędzy 630°C a temperaturą ciekłego helu: 1,92 GPa (rozciągające) dla podłoża ze stali nierdzewnej z powłoką o grubości 13 μm oraz -2,42 GPa (ściskające) dla powłoki $MgO\text{-}ZrO_2$ o grubości 7 μm. Morfologia powierzchni i mikrostruktura próbek zostały również scharakteryzowane za pomocą skaningowego mikroskopu elektronowego (SEM). Obserwacje w SEM wykazały, że powłoki $MgO\text{-}ZrO_2$ mają strukturę pęknięć podobną do mozaiki.

1. Introduction

High temperature $MgO\text{-}ZrO_2$ insulation coatings are deposited on long-length Stainless Steel tapes for magnet technology using reel-to-reel sol gel technique. Ceramic insulation which is vitally essential to prevent electric short circuits within the winding of a coil, is made from E-glass, S-glass, standard quartz by a vacuum impregnation with a resin and ZrO_2 and ZrO_2 based ceramic coating [1, 2].

ZrO_2 based materials due to the corrosion resistance, high hardness, large relative dielectric constant, thermal stability, ionic and electrical conductivity, etc. are suitable to use high temperature insulation and other applications [3, 4]. Therefore ZrO_2 and ZrO_2 based materials have been studied by many researchers using various methods [1-9]. Among these methods the reel-to-reel, continuous sol-gel technique is very useful for insulation coating (details are available in [1, 2, 10, 11]). However, ceramic insulation coatings usually suffer failure cause by flaking and cracking [12] during cooling due to excessive residual stresses generated near the interface and poor bonding strength between $MgO\text{-}ZrO_2$ coating and Stainless Steel (SS) substrate. The critical stresses of the bonded system occur in coatings between substrate and films because of thermal and elastic mismatch effects [13-16]. These effects are relatively substrate coating thickness, thickness of interlayers and fracture resistance of the interface; the plastic flow stress of the metal etc. From the Refs. [10, 11] failures in sol-gel coatings also depend on processing parameters; i.e. solution properties (precursors, solvents, chelating agent, pH value, gelation, viscosity, chelation and complexition rate), withdrawal rate, drying, heat treatment and annealing conditions.

The aim of the present work is to investigate the residual stresses arise for the long length, homogeneous $MgO\text{-}ZrO_2$ insulation coating on Stainless Steel substrate for magnet technology using reel-to-reel sol gel technique.

2. Experimental procedure

The $MgO\text{-}ZrO_2$ solutions and polycrystalline powder samples are synthesized by sol-gel process using Magnesium 2, 4 pentanedionate and Zirconium tetrabutoxide. After Stainless Steel (SS-304) tapes are cleaned using pure acetone the $MgO\text{-}ZrO_2$ coatings are applied on commercial SS tapes by

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using a reel-to-reel sol-gel dipping process with speed of 0.65 m/min by using vertical three-zone furnace. Process is repeated 4, 8, 9 times [1]. The dip coating is composed of dipping, withdrawal, and heating. Coating thickness is important factor for magnitude of stress components [10, 11].

Surface morphology, thickness and stoichiometry of coating films are observed using the Environmental Scanning Electron Microscope (ESEM, electro scan model E-3 and Jeol-5910LV) and the Tencor Alpha-step 200 profilometer.

3. Analysis of residual stresses

The thicknesses of MgO-ZrO_2 coatings are 13, 12 and 7 μm in the top and bottom of the substrate and that of SS is approximately 25 μm. The width of insulation coating to be 0.5 cm is relatively big to its own thickness. The stress components in the middle of insulation coating are defined to be a thick plate with free of surface traction. The area of the cross section of insulation coating is too small. Therefore, distribution of the temperature on its own cross section can be considered to be invariable in the cooling rate applied to it. And, the stresses occurring in insulation coating during the cooling process can be assumed as quasi-static at any time. So, the stress components in the mid of structure can be given as follows:

$$\sigma_{xx} = \sigma_{zz} = f(y) \quad (1)$$

$$\sigma_{yy} = 0,\ \sigma_{xy} = 0,\ \sigma_{yz} = 0,\ \sigma_{xz} = 0 \quad (2)$$

The stress-state given above can be expressed of the following form [17]:

$$\sigma_{xx} = \sigma_{zz} = -\frac{\alpha E}{1-\nu}\Delta T + c_1 + c_2 y \quad (3)$$

where α denotes the thermal expansion coefficient, E represents Young's modulus, ν and ΔT are Poisson's ratio and difference in the temperature, respectively. c_1 and c_2 are integration constants to be determined from the boundary conditions of zero tractions on the edges of the plate. The resultant force and moment (per unit of length) produced by σ_{xx} and σ_{zz} are zero over the edges of the plate for any temperature as shown in Fig. 1, that is,

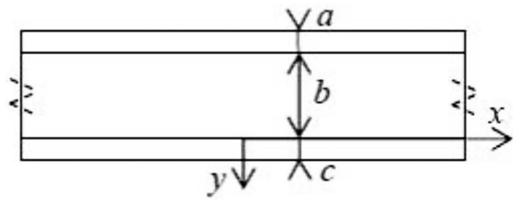

Fig. 1. Investigated section

$$\int_{-(a+b)}^{c} \sigma_{xx} dy = 0 \quad (4)$$

$$\int_{-(a+b)}^{c} \sigma_{xx} y dy = 0 \quad (5)$$

The constants are obtained using the known thicknesses and boundary conditions, Eqs. (4) and (5), as follows:

$$c_2 = \left(-\frac{6\Delta T}{((a+b)^3+c^3+3(a+b)c(a+b+c)}\right)\left(\left(\frac{\alpha_2 E_2}{1-\nu_2}\right)((a+b)c - bc\right.$$

$$\left. + b(a+b) - b^2) - \left(\frac{\alpha_1 E_1}{1-\nu_1}\right)(a+b)c + \left(\frac{\alpha_2 E_2}{1-\nu_2}\right)(b^2 + bc - (a+b)b)\right)$$

$$c_1 = \frac{1}{(a+b+c)}\left[\left(\left(\frac{\alpha_2 E_2}{1-\nu_2}\right)(a+b) + \left(\frac{\alpha_1 E_1}{1-\nu_1}\right)c\right)\Delta T - \right.$$

$$\left. -(((c^2 - (a+b)^2)/2)c_2)\right]$$

where index *1* represents the MgO-ZrO_2 coating, while *2* does the SS substrate.

The stress components for the presented sample are calculated using the formulation given above for four different temperature regimes (Case I to Case IV). The results are presented in the following Figs. 2 to 4.

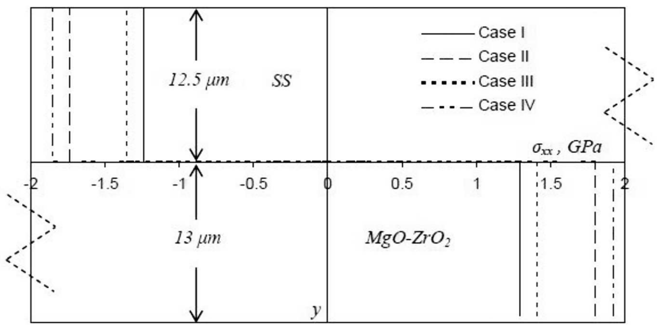

Fig. 2. Variation of stress components versus$y$axis (thickness of MgO-ZrO_2 is 13 μm)

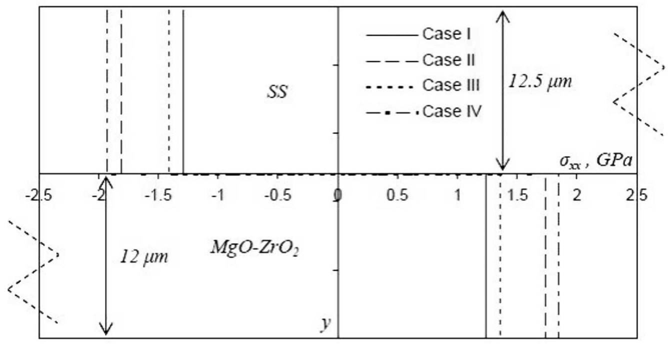

Fig. 3. Variation of stress components versus y axis (thickness of MgO-ZrO_2 is 12 μm)

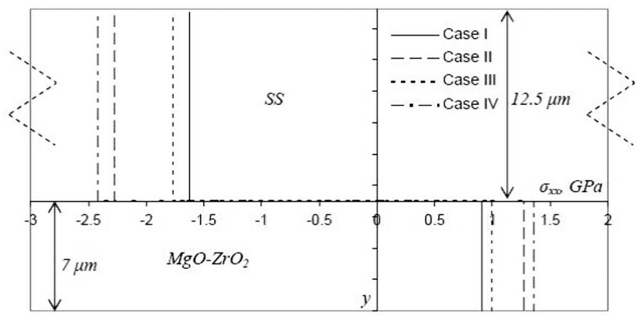

Fig. 4. Variation of stress components versus y axis (thickness of MgO-ZrO_2 is 7 μm)

Using advantage of the symmetry, the only half of the results is given in the figures. And, it is also noted that σ_{xx}

is equal to σ_{zz} while σ_{yy} =0. Cases correspond to the temperatures between 580°C and 25°C, 580°C and -196°C, 630°C and 25°C, 630°C and -196°C, respectively. Material properties of the substrate and coatings are given in TABLE 1 to be analyzed the stress state in the buffer layer structure in room temperature.

It is apparent from Figs. 2 to 4, the stress components have high magnitudes.

TABLE 1

Material properties

	$MgO\text{-}ZrO_2$	SS
Modulus of elasticity (E) (GPa)	28	200
Poisson's ratio (ν)	0.24	0.3
Thermal expansion coefficient (α) ($10^{-6}/K$)	7.8	17

4. Results and discussion

High temperature $MgO\text{-}ZrO_2$ insulation coatings were deposited on SS substrates with various dip numbers by the reel-to-reel sol-gel process. Thickness of $MgO\text{-}ZrO_2$ insulation coatings, uniform along the samples, is determined by SEM which is about 7, 12 and 13 μm for insulation 4, 8 and 9 dips. Thermal stress analysis of $MgO\text{-}ZrO_2$ insulation coating on SS substrates by sol–gel dip coating process is investigated as a function of temperature and thickness. SEM observation indicates that $MgO\text{-}ZrO_2$ coatings have the cracks, pinholes and mosaic structure, desired in ceramic insulators in Figs. 5 and 6. Cracks are also seen in the cross-section in Fig. 6. However these cracks are decreasing with reducing thickness.

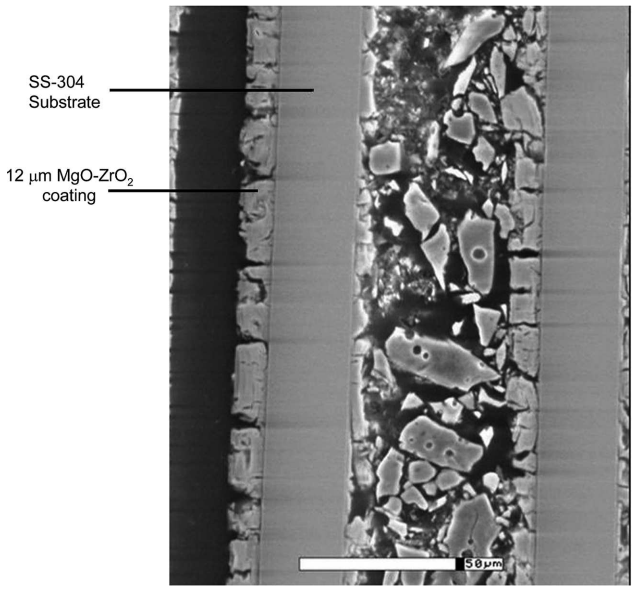

Fig. 5. SEM micrographs of a longitudinal cross-section of 8 dips $MgO\text{-}ZrO_2$ coatings on SS-304 tape (the scale bar is 50 μm)

The stress components ($\sigma_{xx} = \sigma_{zz}$) values for the center and boundary of the substrate (SS) and boundaries of the $MgO\text{-}ZrO_2$ coating, which are the critical points, are shown in Figs. 7-10, respectively for each case. As seen in these figures, stress component values of $MgO\text{-}ZrO_2$ coating are compressive, but stress component values of substrate (SS) are tension in each case. Although variation of stress component ($\sigma_{xx} = \sigma_{zz}$) versus along to thickness is linear in Eq. (3), in the figures they are seen to be constant due to the very thin of the thickness.

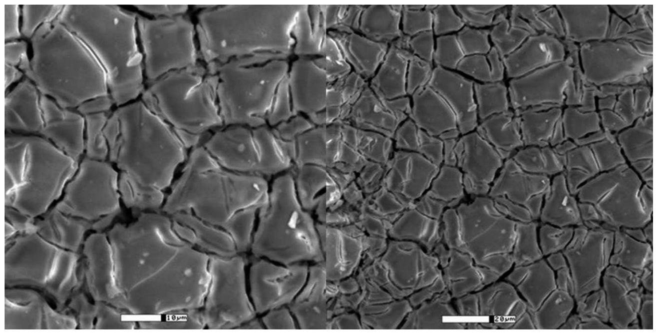

Fig. 6. SEM micrographs of $MgO\text{-}ZrO_2$ insulation on SS-304 tape. The white scale bars are a) 10 μm and b) 20 μm, respectively

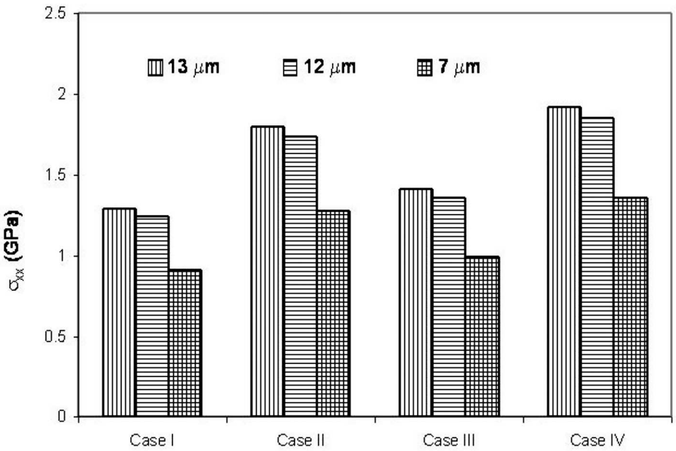

Fig. 7. Stress distribution in the middle of substrate (SS) for different cases

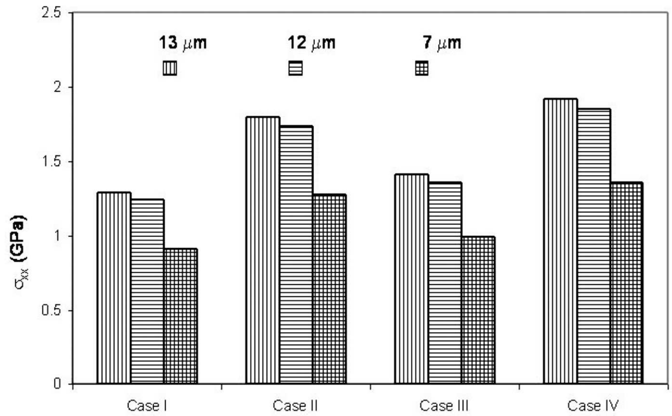

Fig. 8. Stress distribution in the boundary of substrate (SS) for different cases

It is also found that the magnitude of stress is quite high especially between the coating and substrate in the applied temperatures. It is seen that the stress components are constant in each layer with a small discontinuity among layers. The magnitude of the stress component of 13 μm coating thickness is smaller than substrate but they are larger than in

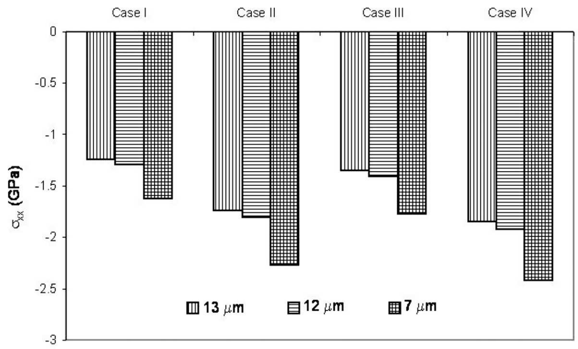

Fig. 9. Stress distribution on the inner boundary of MgO-ZrO_2 coating for different cases

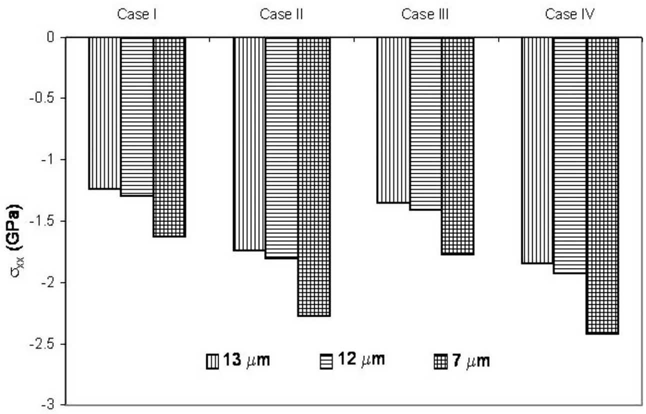

Fig. 10. Stress distribution on the outer boundary of MgO-ZrO_2 for different cases

both of 12 and 7 coating thickness. While the thickness of coating decrease the stress component values of substrate decrease, but the stress values of coating increase. The maximum stress values, which are 1.92 GPa for substrate and -2.42 GPa for the coating in Figs. 7 and 10, are obtained in case IV, which is the temperature range between 630°C-(-196°C). Subtraction of magnitudes of stress components on the interface between substrate and coating are obtained constant independent from the coating thickness.

5. Conclusions

The high temperature MgO-ZrO_2 coatings on SS substrates are fabricated by the reel-to-reel sol-gel process. SEM observation indicates that MgO-ZrO_2 coatings have the cracks and pinholes which occur from solution prepared from alkoxide precursor, solvent and chelating agent. Thermal stress analysis of MgO-ZrO_2 insulation coating on SS substrates by sol–gel dip coating process is investigated as a function of temperature and thickness using analytical method. Especially the interface between the coating and substrate is found critical in the applied temperatures for the magnitude of stress components. It is also seen that the stress components are constant in each layer with a small discontinuity among the layers.

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REFERENCES

[1] O. Cakiroglu, L. Arda, Y. S. Hascicek, Physica C **422**, 117 (2005).
[2] H. Mutlu, Y.S. Hascicek, U.S. Patent Number: 6,344,287 issued 2/5/02.
[3] A. Danelska, M. Szafran, E. Bobryk, Archv. Metall. Mater. **54**, 1029 (2009).
[4] E. Iller, D. Wawszczak, L. Gorski, Archv. Metall. Mater. **57**, 443 (2012).
[5] T. Olding, M. Sayer, D. Barrow, Thin Solid Films **398**, 581 (2001).
[6] G. Carta, N. El Habra, G. Rossetto, P. Zanella, M. Casarin, D. Barreca, C. Maragno, E. Tondello, Surf. Coat. Tech. **201**, 9289 (2007).
[7] P. Ctibor, J. Sedlacek, K. Neufuss, Ceram. Int. **29**, 527 (2003).
[8] B.Q. Liu, F.L. Qin, J.L. Jiang, Z.J. Jin, J. Mater. Eng. Perform. **21**, 308 (2012).
[9] M. Yasmeen, S.A. Ansar, A.A. Khan, T. Joyeux, J.C. Labbe, J. Mater. Eng. Perform. **10**, 542 (2001).
[10] L. Arda, S. Ataoglu, J. Alloy Compd. **471**, 282 (2009).
[11] L. Arda, S. Ataoglu, S. Sezer, Z. Abdulaliyev, Surf. Coat. Tech. **202**, 439 (2007).
[12] J.H. Kim, J.B. Song, Y.H. Jeong, Y.J. Lee, J.H. Paik, W.S. Kim, H. Lee, Met. Mater. Int. **16**, 109 (2010).
[13] F. Yang, W.D. Fei, Z.M. Gao, J.Q. Jiang, Surf. Coat. Tech. **202**, 121 (2007).
[14] J. Peng, V. Ji, W. Seiler, A. Tomescu, A. Levesque, A. Bouteville, Surf. Coat. Tech. **200**, 2738 (2006).
[15] B. Girault, P. Villian, E.L. Bourhis, Surf. Coat. Tech. **201**, 4372 (2006).
[16] H. Okuda, H. Rokkaku, K. Morishita, J.K. Shin, S. Iwamoto, S. Ochiai, M. Sato, K. Osamura, A. Otto, E.J. Harley, A. Malozemoff, Scripta Materialia **55**, 691 (2006).
[17] B.A. Boley, J.H. Weiner, Theory of Thermal Stresses, Wiley, New York (1960).

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