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Fabrication of biaxial textured NiO on Ni in a one-step process

Research Article

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Abstract: A novel method for the preparation of biaxial textured nickel oxide on commercially available nickel via a modified surface oxidation epitaxy (SOE) process has been developed. Following studies of different heat-treatment procedures for both texturing of nickel and for the fabrication of nickel oxide the following method was found to yield the best results. Nickel was first textured under an argon – hydrogen atmosphere at 1000°C for 120 min, then the temperature was lowered to 800°C and the atmosphere was changed to argon with 3 ppm oxygen. Smooth and crack free c-axis textured and a-b aligned NiO buffer layers with an out-of-plane texture of 7.8° and an in-plane texture of 9.4° were successfully produced. Higher oxygen partial pressure and temperatures resulted in increased surface roughness and excessive grain growth.

Keywords: *Coated conductors • NiO • Surface oxidation epitax • Buffer layer*

1. Introduction

The second generation of high temperature superconducting (HTSC) tapes is based on coated conductor technology. Cube textured Ni and Ni alloys prepared by the RABiTS (rolling assisted biaxial textured substrates) method [1] are widely used substrates for coated conductors. Many substrate properties such as texture sharpness, mechanical and thermal stability, magnetic behavior and solution hardening depend on the quality of Ni [2].

Coated conductors consist of (textured) metal substrates covered with one or more buffer layers. Buffer layers are necessary to suppress the diffusion of metal atoms from the substrate to the superconducting phase. They also block the diffusion of oxygen to the metal tape, limiting oxidation of the substrate. In addition the buffer layers (e.g. $Y_xZr_{1-x}O_{2-\delta}$, CeO_2, $LaMnO_3$, $SrTiO_3$, $BaZrO_3$, NiO) must promote texture of the superconducting layer.

The superconducting weak coupling at the grain boundary is a serious problem in $REBa_2Cu_3O_{7-x}$ materials [3]. The important j_c values for HTSC-tapes are significantly decreased when the mis-orientation angle between the grains exceeds 10° [4]. Thus both substrates and buffer layers must show mis-orientation angles smaller than 10° in order to permit high critical current densities in the superconducting layer.

NiO buffer layers are generally prepared by the surface oxidation epitaxy (SOE) method [5-7]. This procedure characterized by the controlled epitaxial growth of NiO on textured Ni or Ni alloys. An advantage of this method is the lower cost of production compared to pulsed laser deposition (PLD) techniques. An intermediate treatment between recrystallization of the metal tape and the SOE process is necessary to remove the initial oxide layer built up during storage of Ni [8-10]. Detailed growth conditions for the formation of textured NiO layers have been examined by several groups [11-14].

The production, the properties and the potential of Ni tapes prepared from commercially available material have been reported elsewhere [15]. The main goal of this work is to examine commercial nickel substrates for the SOE process. In addition we report a combined texturing and oxidation process which makes an intermediate treatment between recrystallization of the tapes and controlled oxidation unnecessary.

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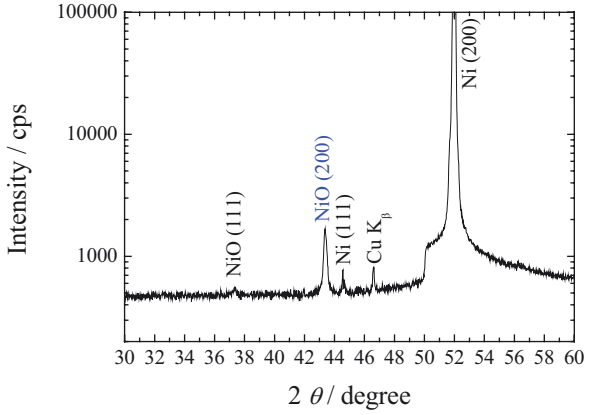

Figure 1. X-ray diffraction pattern of NiO on a textured Ni substrate formed at 800°C under argon with 3 ppm oxygen. Ni – filtered Cu K_α radiation.

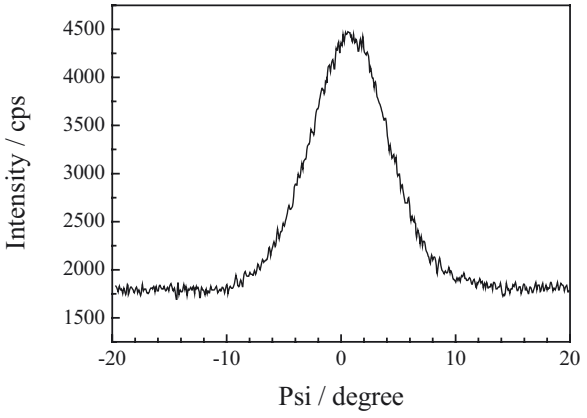

Figure 2. Ψ - Scan of the (200) peak of NiO on a textured Ni substrate formed at 800°C under argon with 3 ppm oxygen (FWHM: 7.8°).

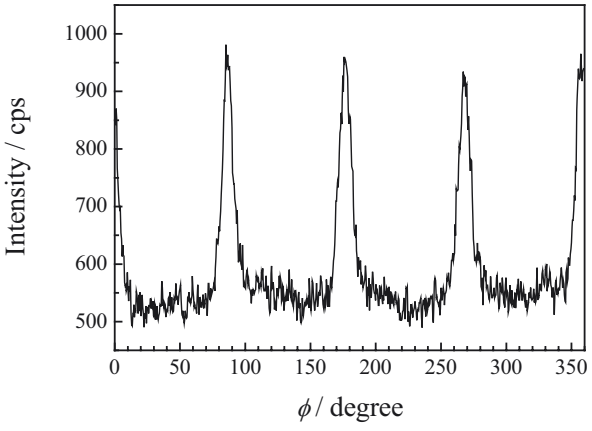

Figure 3. ϕ scan of the (111) peak of NiO on a textured Ni substrate formed at 800°C under argon with 3 ppm oxygen (FWHM: 9.4°).

2. Experimental Procedures

Detailed information for the fabrication of tapes prepared from commercially available Ni (98% purity) has already been published [15]. The cold rolled Ni tapes, approximately 50 cm long and 1 cm wide, were ultrasonically cleansed with acetone and methanol to remove roller oil and other impurities. Samples of 10 mm x 10 mm were cut from the tape, placed in an alumina crucible and transferred into a tube furnace. In the first step of the process the Ni tapes were heated to achieve cubic biaxial texture. The furnace was evacuated and then filled with a reducing argon-hydrogen gas mixture (6.5% vol. - H_2). Heating rates to the peak temperature varied between 3 and 10 K min^{-1}. Final temperatures between 800°C and 1300°C were chosen, over durations ranging from 15 to 300 min. In the next step the atmosphere in the furnace was changed by introducing either air or argon with a small oxygen content (approximately 3 ppm) for surface oxidation of nickel. The whole system was evacuated with a turbo-molecular pump. The pressure for either air or argon as atmosphere was varied between 10^{-6} and 1 bar during this study. The furnace temperature was varied between 800°C and 1300°C, with durations of the oxidation ranging between 10 minutes and 2 hours.

An X`Pert Pro (Panalytical) diffractometer was used for all X-ray measurements employing Ni-filtered Cu K_α radiation. Texture measurements were made with the help of an Eulerian cradle. Scanning electron micrographs were obtained with a JSM-6400 (Jeol, Japan).

3. Results and Discussion

The best conditions for preparing smooth, coherent NiO films were as follows: The Ni tape was heated at 1000°C for 120 min in Ar + 6.5 vol-% H_2. The tube of the furnace was then evacuated and the furnace was cooled at a rate of 3 K min^{-1} to 800°C. The tube of the furnace was first flushed with flowing argon containing 3 ppm O_2. A pressure of 10^{-2} mbar was then applied during the oxidation while maintaining a slow flow of argon with traces of oxygen through the system.

An X-ray diffraction pattern of a textured NiO layer on Ni prepared at 800°C under argon with 3 ppm oxygen is shown in Fig. 1. A logarithmic presentation of the intensity was chosen to enhance possible low intensity peaks, especially the (111) peak of NiO. The XRD pattern showed the (200) peak of NiO, the (111) peak is barely visible. This confirms that excellent *c*-axis orientation of the NiO layer has been obtained.

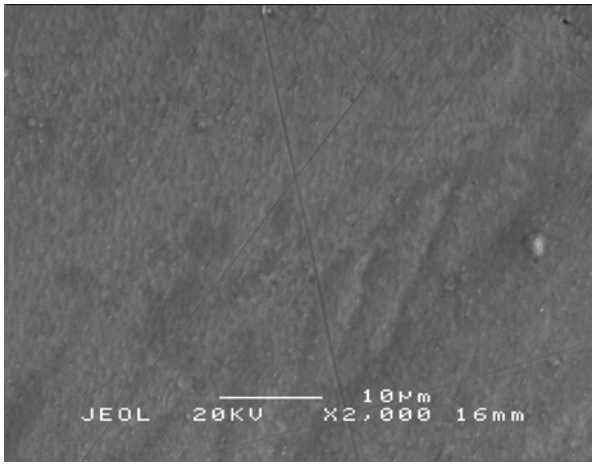

Figure 4. Scanning electron micrograph of the surface of NiO layer on Ni tape prepared at 800°C under reduced oxygen partial pressure.

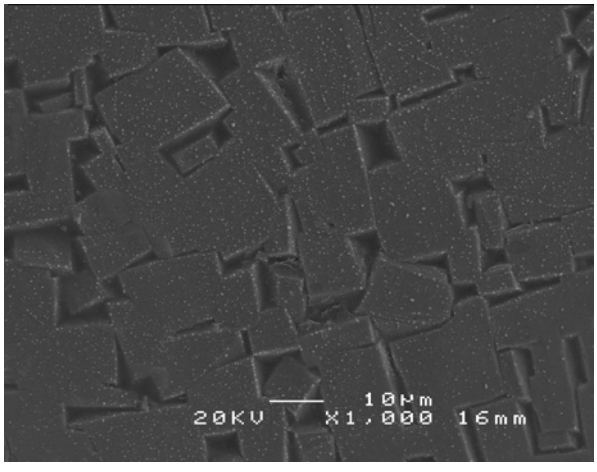

Figure 5. Scanning electron micrograph image of the surface of the NiO layer on Ni tape prepared at 1300°C under air.

The FWHM for the rocking curve (Ψ - scan) of the (200) peak (2θ = 43.3°) of NiO was 7.8° (Fig. 2), the FWHM for the ϕ - scan measured on the (111) peak of NiO (2θ = 37.2; Ψ = 54.7°) was 9.4° (Fig. 3). Thus, as well as excellent *c*-axis texturing, also outstanding *a-b* alignment of the NiO layer has been achieved.

The surface properties of cubic biaxial textured NiO layers depend strongly on the oxidation procedure. The comparison between the scanning electron micrographs of NiO surfaces (Fig. 4 and 5) oxidized under different conditions shows the advantage of the modified method developed during this study. NiO buffer layers created at lower temperatures and lower oxygen amounts (Fig. 4) showed smoother surfaces without cracks compared to the pronounced cubes observed in samples which were oxidized in air at 1300°C in ambient pressure (Fig. 5). These buffer layers had cubic crystallites with edge length of about 10 μm. Additionally NiO layers

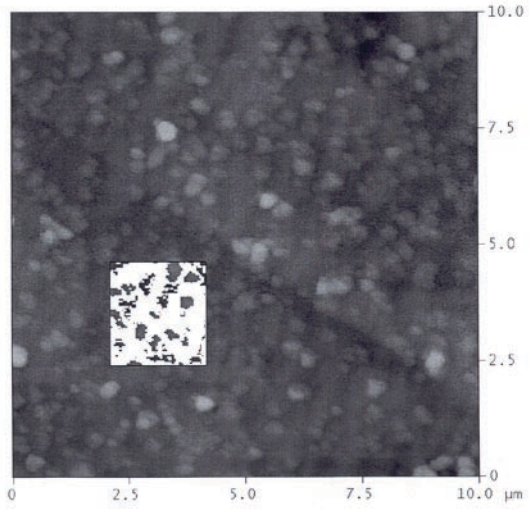

Figure 6. Two-dimensional atomic force microscopy image (10 μm x 10 μm) of a NiO buffer layer oxidized at 800°C.

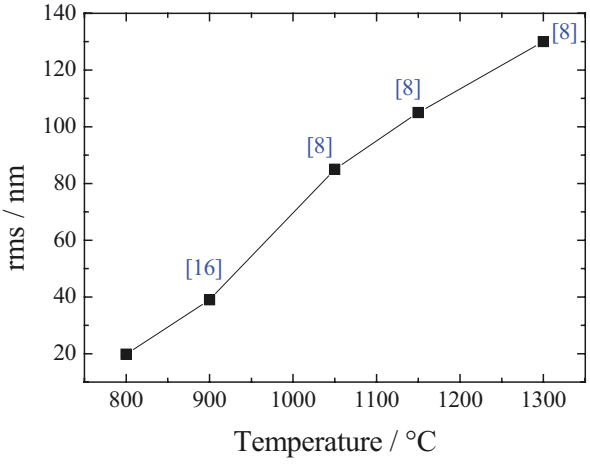

Figure 7. Surface roughness (rms) dependency on oxidation temperatures. Numbers in brackets refer to literature data.

as shown in Fig. 5 had slightly worse FWHM values in the *a-b*-plane of 10°. Therefore one may expect that superconducting layers applied to such NiO layer will probably show lower j_c values.

We determined the surface roughness of the most promising NiO layer prepared by the novel self-oxidation process at 800°C under reduced oxygen partial pressure with atomic force microscopy. The scan area of the unpolished NiO was set to 10 x 10 μm² (Fig. 6). The AFM analysis yielded a roughness mean square value (rms) of the NiO layer of less than 20 nm (19.833 nm). The bright marked box has rms value of 11.690 nm and does not incorporate the grain boundary in the middle of the section.

R. Hühne *et al.* [8] report rms values of SOE layers on Ni-Cr tapes of 85 nm oxidized at 1050°C, 105 nm oxidized at 1150°C and 130 nm at 1300°C. J.-K. Chung *et al.* [16] describe the epitaxial growth of NiO on bi-

axially textured Ni-3 at.-%W substrates by line-focused infrared heating. Their optimal oxidation conditions were 900°C and the buffered tapes had rms roughness values of 39 nm (AFM: 10 x 10 μm). Our results are better than those published and support our approach that oxidation at lower temperatures results in smoother NiO buffer layers (Fig. 7).

4. Conclusions

Commercially available Ni (98.5% purity) was successfully used as substrate for the formation of a textured NiO buffer layer by a SOE process. The novel self oxidation process enabled integrated oxidation of Ni directly after texturing of the metal substrate to form textured NiO buffer layers without any intermediate treatment. Best results were achieved with the oxidation in flowing Ar with 3 ppm oxygen at 800°C at 10^{-2} mbar. These NiO buffer layers showed out-of-plane alignment of 7.8° and in-plane alignment of 9.4° and had a smooth surface (rms < 20 nm). The advantages of this method are the lower cost of the raw material, the elimination of intermediate treatment steps and the relatively moderate oxidation temperature.

Acknowledgments

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References

[1] D.P. Norton et al., Science 274, 755 (1996)
[2] T.G. Woodcock, J.S. Abell, T.C. Shields, M.G. Hall, Physica C 372-376 (SUPPL. 2), 863 (2002)
[3] T.G. Woodcock, J.S. Abell, M.G Hall, J. Microscopy 205, 231 (2002)
[4] T. Watanabe, K. Matsumoto, T. Maeda, T. Tanigawa, I. Hirabayashi, Physica C 357-360 (SUPPL. 1), 914 (2001)
[5] K. Masumoto et al., IEEE Trans. Appl. Supercond. 9, 1539 (1999)
[6] C.F. Liu et al., IEEE Trans. Appl. Supercond. 9, 1471 (1999)
[7] A. Kursumovic et al., Physica C 385, 337 (2003)
[8] R. Hühne et al., J. Phys. D Appl. Phys. 36, 1053 (2003)
[9] M.J. Graham, M.J. Cohen, Electrochem. Soc. 120, 879 (1972)
[10] M.J. Graham, R.J. Hussey, M.J. Cohen Electrochem. Soc. 120, 1523 (1973)
[11] K. Matsumotoet al., Physica C 335, 39 (2000)
[12] Z. Lockman, X. Qi, A. Berenov, R. Nast, W. Goldacker, J.MacManus-Driscoll, Physica C 351 34 (2001)
[13] J. Yang et al., Physica C 412–414, 844 (2004)
[14] A. Kursumovic, R. Hühne, R. Tomov, B. Holzapfel, B.A. Glowacki, J.E. Evetts, Physica C 405, 219 (2004)
[15] O. Staller, D. Holzmann, G. Gritzner, P. Diko, D. Mikolaj, F. Kováč, Cent. Eur. J. Chem. (accepted)
[16] J.-K. Chung, W.-J. Kim, J. Tal, C.J. Kim, Physica C 463-465, 619 (2007)