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Phase equilibria of the Ba–Sm–Y–Cu–O system for coated conductor applications

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ABSTRACT

The complex phase relationships near the BaO-poor region of the quaternary Ba–Sm–Y–Cu–O oxide system prepared in pure air ($p_{O_2}=22$ kPa, 950 °C) and in 0.1% O_2 ($p_{O_2}=100$ Pa, 810 °C) have been determined. This investigation also included the subsolidus compatibilities in ten subsystems (Ba–Sm–Y–O, Ba–Sm–Cu–O, Ba–Y–Cu–O, Sm–Y–Cu–O, Ba–Sm–O, Ba–Y–O, Ba–Cu–O, Sm–Y–O, Sm–Cu–O, and Y–Cu–O), and the homogeneity range of five solid solutions ($Ba(Sm_xY_{2-x})CuO_5$, $(Sm,Y)_2O_3$, $(Sm,Y)_2CuO_4$, $(Y,Sm)_2Cu_2O_5$, and $Ba(Sm,Y)_2O_4$). The single phase range of the superconductor solid solution, $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$, and the phase compatibilities in its vicinity, which are particularly important for processing, are described in detail. The phase equilibrium data of the Ba–Sm–Y–Cu–O system will enable the improvement of the intrinsic superconducting properties of second-generation wires, and facilitate the flux-pinning process.

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1. Introduction

Current economic and environmental issues demand improvements in electrical distribution grids for more efficient utilization of energy resources. High-temperature superconductors have demonstrated potential for meeting these needs [1]. There is continued effort within the high T_c community on research and development of coated conductors for wire and tape applications [2–7]. These coated conductors are based on $Ba_2YCu_3O_{6+z}$ (Y-213) and $Ba_2RCu_3O_{6+z}$ (R-213, R=lanthanides) as the principal superconductors. Y-213 and R-213 can be deposited on flexible substrates using various approaches, including the BaF_2 technique [8–14]. The resulting tapes show excellent current-carrying capability. Phase equilibrium data on the multi-component phase relationships of mixed lanthanide high temperature superconductor phases are important for coated conductor wire/tape processing.

Since an understanding of the detailed multi-component phase equilibrium relationships in the vicinity of the superconductor solid solutions will facilitate optimization of coated conductor processing, our goal is to provide critical data for these single phase solid solutions. Such data will enable improvement of the intrinsic superconducting properties of second-generation wires and the associated flux-pinning processes. For example, one can tailor superconducting and melting properties of the mixed lanthanide 213 solid solutions by substitution of larger lanthanides on the Ba site, and also by mixing larger lanthanide ions (R) with smaller ones (R'). MacManus-Driscoll et al. reported that compositions in the $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$ system give rise to enhanced J_c properties [15]. The compatibilities of this solid solution with its neighboring phases are crucial as a reference for coated conductor processing as well as for property investigations.

This paper reports the complex phase equilibria of the Ba–Sm–Y–Cu–O system as well as the single phase solid solution region of $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$. Since X-ray patterns are important to serve as standards for phase analysis, we have also prepared reference X-ray powder diffraction patterns of selected solid solutions in the $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$ and $Ba(Sm_xY_{2-x})CuO_5$ systems to be included in the Powder Diffraction File (PDF, produced by the International Centre for Powder Diffraction (ICDD) [16]).

2. Experimental[1]

2.1. Sample synthesis

Ten end members that constitute various multi-component regions in the Ba–Sm–Y–Cu–O system were synthesized and used as master batches for preparing nine individual batches of $Ba_2SmCu_3O_{6+z}$, $Ba_2YCu_3O_{6+z}$, $BaSm_2CuO_5$ (Sm-121), BaY_2CuO_5 (Y-121), $BaSm_2O_4$, BaY_2O_4, Sm_2CuO_4, $Y_2Cu_2O_5$, and $BaCuO_2$. These

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master batches were prepared by heat-treating a stoichiometric mixture of $BaCO_3$, Sm_2O_3, Y_2O_3, and CuO. Samples were weighed out with < 1.0% (relative) estimated uncertainty for component compositions of 10–100 mol%, and < 5% (relative) estimated uncertainty for component compositions with less than 10 mol%. Following this, samples were well-mixed and calcined first at 850 °C, followed by final annealing at 900–1000 °C for about 15 days with intermediate grindings to ensure single phase properties. Samples were prepared using different combinations of these nine master batches as well as using Y_2O_3, Sm_2O_3, and CuO, depending on the equilibrium regions of the Ba–Sm–Y–Cu–O system under investigation (tables listing the 274 compositions investigated in this study have been deposited as Supplementary Tables (Tables S1–S3)). For samples prepared in air, the heat-treatment procedure was: 850 °C for 20 h, 900 °C for 20 h, 930 °C for 20 h, and 950 °C for 100–400 h with intermediate grindings. For samples prepared in $p_{O_2} = 100$ Pa, samples were heat-treated at 810 °C for 40–200 h with intermediate grindings.

To study the range of the solid solution $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$, two sets of 75 samples were prepared using high temperature solid state methods in $p_{O_2} = 100$ Pa (810 °C for 40–200 h) and in pure air (950 °C for 100–400 h) with intermediate grindings.

The experimental conditions used in this study were selected in consultation with an industry/university/government advisory group to correspond to processing parameters employed by those most active in developing high T_c products for wire and cable applications.

2.2. Phase identification using X-ray diffraction

X-ray powder diffraction was used to identify the phases synthesized, to confirm phase purity, and to determine phase relationships. A computer-controlled automated Philips diffractometer equipped with a θ-compensation slit and Cu$K\alpha$ radiation was operated at 45 kV and 40 mA. The radiation was detected by a scintillation counter and a solid-state amplifier. All X-ray patterns were measured using a hermetic cell designed for air-sensitive materials [17]. The Siemens software package and the reference X-ray diffraction patterns of the Powder Diffraction File (PDF) [16] were used for performing phase identification.

2.3. Structural studies and reference patterns

To study the structure and prepare X-ray reference patterns, the X-ray Rietveld refinement technique (GSAS Suite) [18,19] was applied (neutron diffraction was not used because of neutron absorption by Sm). The structures of selected $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$ and $Ba(Sm_xY_{2-x})CuO_5$ phases were determined using a Bruker XRD D8 Diffractometer equipped with a VANTEC-1 position-sensitive detector. Diffraction patterns were collected (Cu$K\alpha$ radiation, 40 kV, 40 mA, 0.3° divergence slit) from 8° to 140° 2θ using Ni-filtered Cu$K\alpha$ radiation, and counting for 0.3 s/step. The specimens were mounted as acetone slurries on zero-background cells, and were rotated rapidly during data collection. The background was described by a 6-term shifted Chebyshev function of the first kind.

Reference X-ray patterns of selected members of the $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$ and $Ba(Sm_xY_{2-x})CuO_5$ systems were prepared using a Rietveld pattern decomposition technique. These patterns represent ideal specimen patterns. They are corrected for systematic errors in both *d* and *I*. The reported peak positions are calculated from the refined lattice parameters, as these positions represent the best measure of the true positions. For peaks resolved at the instrument resolution function, the individual peak positions are reported. For overlapping peaks, the intensity-weighted average peak position is reported with multiple indices. For marginally resolved peaks, individual peaks are reported to more accurately simulate the visual appearance of the pattern.

3. Results and discussion

The phase relations in the vicinity of BaO poor region of the Ba–Sm–Y–Cu–O system under both pure air ($p_{O_2} = 22$ kPa)

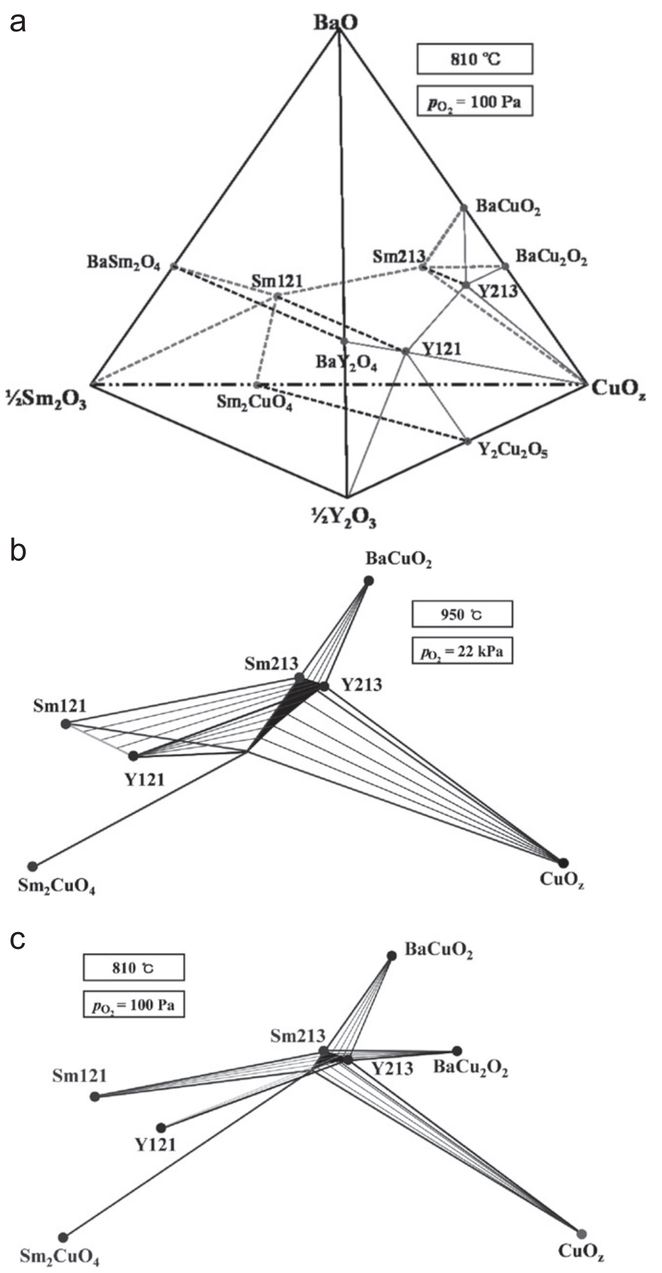

Fig. 1. (a) Phase diagram overview of the Ba–Sm–Y–Cu–O system in the BaO-poor region prepared in $p_{O_2} = 100$ Pa, 810 °C (overview prepared at $p_{O_2} = 22$ kPa, 950 °C is similar, without the presence of $BaCu_2O_2$), (b) schematic illustrating the tie line relationships in the vicinity of the $(Ba_{2-x}Sm_x)(Sm_{1-y}Y_y)Cu_3O_{6+z}$ solid solution at $p_{O_2} = 22$ kPa, 950 °C, and (c) schematic at $p_{O_2} = 100$ Pa, 810 °C.

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[1] The purpose of identifying the equipment in this article is to specify the experimental procedure. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology.