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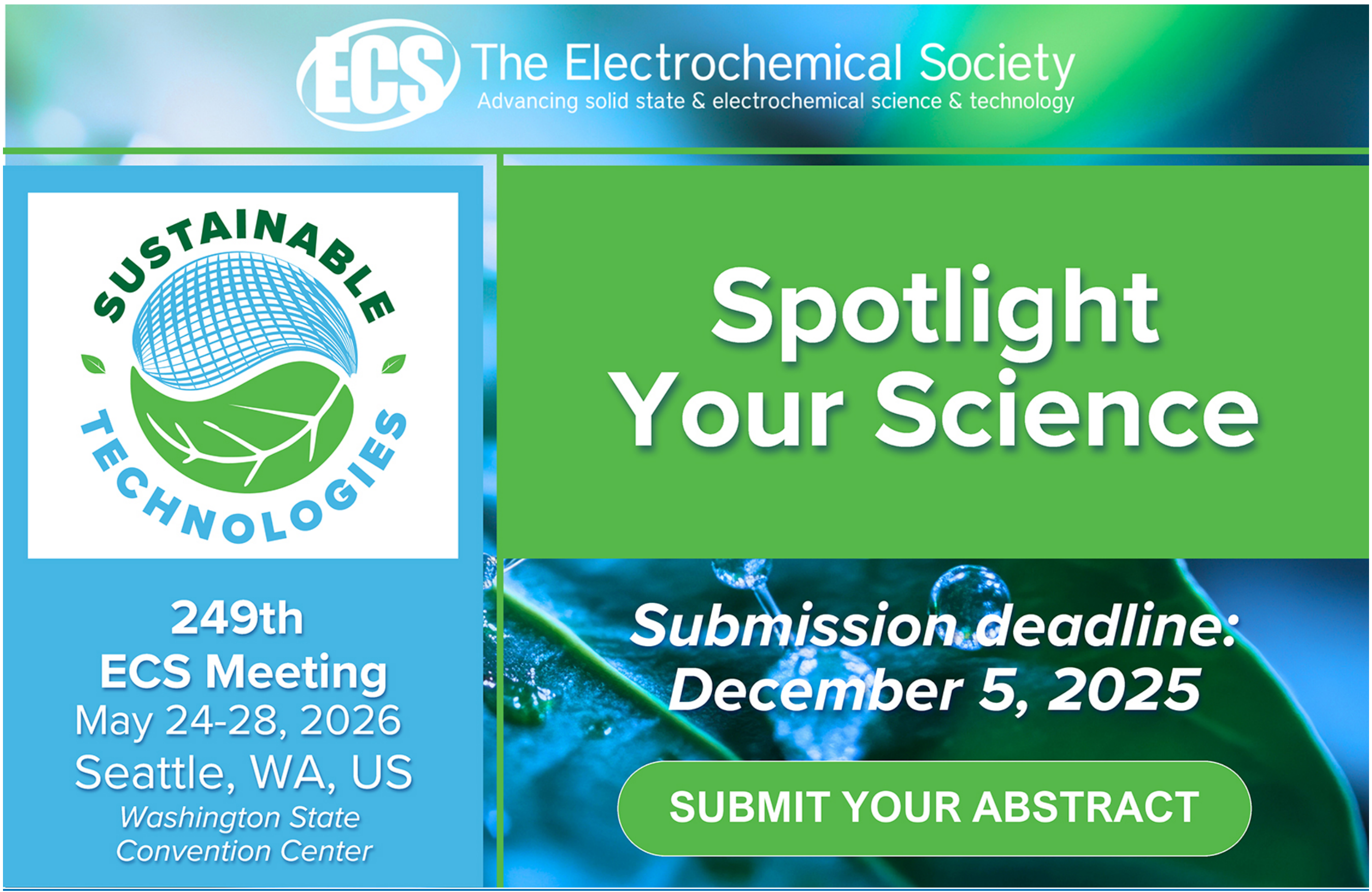

Effect of viscosity of the dispersed media on the bi-axial orientation degrees in magnetically aligned Y123 powder

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Abstract. We clarified the dependence of biaxial orientation degrees on the initial viscosity (η_{init}) and curing time (t_{cure}) of resins used for $YBa_2Cu_3O_y$ (Y123) powder samples aligned under the modulated rotating magnetic fields (MRFs) in two different types of resin. The biaxial orientation degrees of the magnetically aligned Y123 powder samples significantly improved when using a resin with relatively lower η_{init} and a longer t_{cure} within the MRF range of 0.8 – 5 T. These findings contribute to the fabrication of rare-earth-based cuprate superconductor ceramics with high biaxial orientation degrees using permanent magnets and the colloidal process.

1. Introduction

The cuprate superconductor $REBa_2Cu_3O_y$ (RE: Rare Earth elements, RE123) with $y \sim 7$ has a critical temperature (T_c) in the 90 K range [1], which is higher than the boiling point of liquid nitrogen ($\sim$ 77 K). Additionally, RE123 exhibits a high critical current density (J_c) under a magnetic field. Its crystal structure of RE123 consists of alternating layers: a one-dimensional CuO-chain as a blocking layer and two-dimensional CuO_2 plane as the superconducting layer, oriented along the *c*-axis. This anisotropic structure results in a strong directional dependence of J_c, where $J_c//c < J_c//ab$ [2]. The short coherence length and *d*-wave superconducting gap symmetry of the Cooper pair in RE123 lead to serious degradation of the inter-grain J_c. Even in the *c*-axis aligned bicrystals, the inter-grain J_c at a grain boundary with a misorientation angle of $\sim$ 10 degrees is approximately an order of magnitude lower than the intragranular J_c [3]. To achieve high J_c in both self-field and applied-field conditions, it is essential to form a biaxially aligned grain structure and develop a densified microstructure. Therefore, the fabrication of biaxially aligned RE123 materials currently relies on epitaxial growth techniques, such as thin-film deposition [4] and melt-solidification processes [1].

Magnetic alignment using a modulated rotating magnetic field (MRF) [5-9] is a triaxial grain alignment process for materials with triaxial magnetic anisotropies. A key advantage of this method is that it operates at room temperature. Figure 1 shows a schematic of the intermittent type MRF [8], which is the most common configuration. For a superconducting solenoidal magnet with a room temperature bore [10], the MRF is generated by precisely controlling the rotation of the sample within a static magnetic field (B_a) of up to 10 T level. As shown in Fig. 1, the sample remains stationary at 0° and 180°, while it rotates at intermediate angles. In this setup, the nearly linear static magnetic flux lines inside the superconducting magnet bore are perpendicular to the

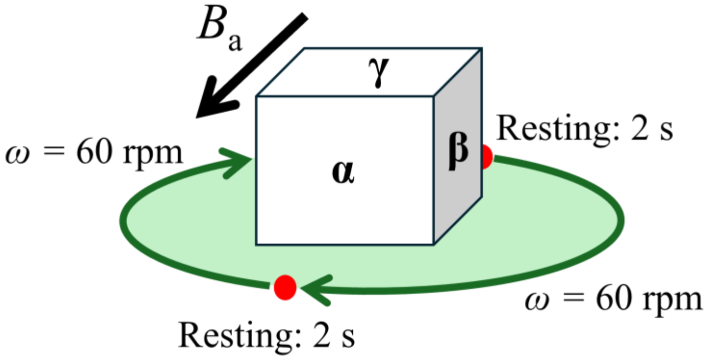

Figure 1. Schematic of the modulated rotating magnetic field of superconducting magnet (SC-MRF). The first easy, second easy, and hard axes of magnetization are aligned perpendicular to the α-, β-, and γ-planes of the magnetically aligned powder samples, respectively.

α-plane at 0°. The magnetic field's rotation plane is configured parallel to the γ-plane. Through this process, the first easy and hard magnetization axes align parallel to the static B_a direction and perpendicular to the rotating field plane, respectively. In principle, for a triaxially magnetically aligned sample, the first easy, second easy, and hard magnetization axes are aligned perpendicular to the α-, β-, and γ-planes, respectively. In 2018, our group developed a linear drive-type MRF (LDT-MRF) apparatus for a continuous process triaxial alignment by using a permanent magnet array [11,12]. From the perspective of cost efficiency, LDT-MRF is preferable, as it enables a continuous process and utilizes permanent magnets, significantly reducing the cost of the magnetic alignment process for RE123. However, RE123 must be aligned under a magnetic field of less than 1 T when using permanent magnets.

To achieve triaxial magnetic alignment, the magnetic orientation energy must be sufficiently high compared to thermal energy, and the magnetic alignment time (relaxation time, τ) must be significantly shorter than the curing or casting time of the resin or colloidal solution. The value of τ depends on the B_a, triaxial magnetic anisotropy ($\Delta\chi$), and viscosity (η) of the dispersed medium [13]. In detail, τ is proportional to η and inversely proportional to $\Delta\chi$ and B_a^2. Utilizing MRF in combination with the colloidal process is essential for fabricating biaxially aligned RE123 ceramics with improved intergranular connectivity. Generally, the viscosity of colloidal solutions used in this process is 100 - 1000 times lower than that of epoxy resin. Therefore, for practical use, it is crucial to clarify the magnetic alignment behavior in dispersed media with lower η levels (~ 0.01 Pa·s).

Among RE123 compounds, $YBa_2Cu_3O_y$ (Y123) is preferred for practical applications due to its relatively lower material cost compared to RE123 with heavy RE ions. However, Y123 has the lowest $\Delta\chi$ in RE123 [14]. Previous studies have shown that biaxial magnetic alignment of Y123 powders in an epoxy resin was not achieved under a 1 T MRF [9], likely because the low $\Delta\chi$ results in an extended τ, which may exceed the curing time of the epoxy resin. Since τ is influenced by η of the dispersed medium, it can be controlled by adjusting η. In the present study, to identify key factors for fabricating magnetically biaxial-aligned Y123 ceramics with high orientation degrees, we investigated the relationship between the dispersed medium and the biaxial orientation degrees of magnetically aligned Y123 powder samples. Biaxial magnetic alignment experiments were conducted on Y123 (y ~ 7) powders under MRFs with varying B_a values in dispersed media with two different initial viscosities (η_{init}) and curing times (t_{cure}). The feasibility of magnetically biaxial-aligned Y123 ceramics was evaluated using (103) pole figures at the α-plane.

Table 1. Details of the initial viscosities (η_{init}) and the curing times (t_{cure}) for Resin A and Resin B.

Type of resin	Initial viscosity (η_{init}) (Pa·s)	Curing time (t_{cure}) (h)
Resin A	40	6
Resin B	0.5	10

2. Experimental details

Y123 polycrystals were synthesized using a standard solid-state reaction in air. The starting materials, Y_2O_3, $BaCO_3$, and CuO were weighed in a cationic ratio of Y: Ba: Cu = 1:2:3 and thoroughly ground in ethanol. The mixture was calcined twice at 880 and 900°C, with intermediate grinding. It was then pelletized and sintered at 920°C for 24 h in air [15]. The obtained Y123 polycrystals were annealed at 300°C in flowing oxygen gas to achieve $y \sim 7$, then pulverized in an agate mortar to obtain powders with an average grain size of ∼ 5 μm. The pulverized Y123 powders were mixed with epoxy resins at a weight ratio of powder to resin = 1:10 and aligned under MRFs (see Fig. 1) with B_a = 0.8, 1, 3, 5, and 10 T for over 12 h at room temperature. In the present study, the resting time and the rotation speed of MRF were set to be 2 s and 60 rpm, respectively. To determine the biaxial orientation degrees, pole figures of the (103) plane at α-plane were examined on fully cured resins containing the magnetically aligned Y123 powders. The biaxial orientation degree (F) was calculated using the (103) pole figure and the following formula,

$$F\ (\%) = \frac{I_{aligned}}{I_{all}} \times 100 \tag{1}$$

where I_{all} is the summation of the intensities in a whole measurement region, and $I_{aligned}$ is the summation of the intensities at four peaks, including intensities within 5° in radius from the four peaks at $\Psi \sim 45°$. In principle, perfect biaxial alignment leads to F = 100 %. However, due to the background intensities in realistic pole figure measurements, F does not achieve 100 % even in the perfect biaxial aligned sample. In the present study, F is used as a relative index on the biaxial orientation degrees.

3. Results and discussion

Table 1 shows the details of the initial viscosities (η_{init}) and the curing times (t_{cure}) for Resin A and B, determined experimentally using viscosities measurement equipment. The initial viscosities of Resin A and B were approximately 40 and 0.5 Pa·s, respectively. A preliminary study by our group found that the magnetic alignment of Dy123 was achievable even in epoxy resins with $\eta_{init} \sim 1000$ Pa·s [16]. Based on this finding, in the present study, the curing time (t_{cure}) of the epoxy resin was conventionally defined as the time at which η reaches 10^5 Pa·s. The measured t_{cure} values were approximately 6 h and 10 h for Resin A and B, respectively. In summary, Resin A exhibited a higher η_{init} and a shorter t_{cure} than Resin B, while Resin B had a lower η_{init} and a longer t_{cure} compared to Resin A.

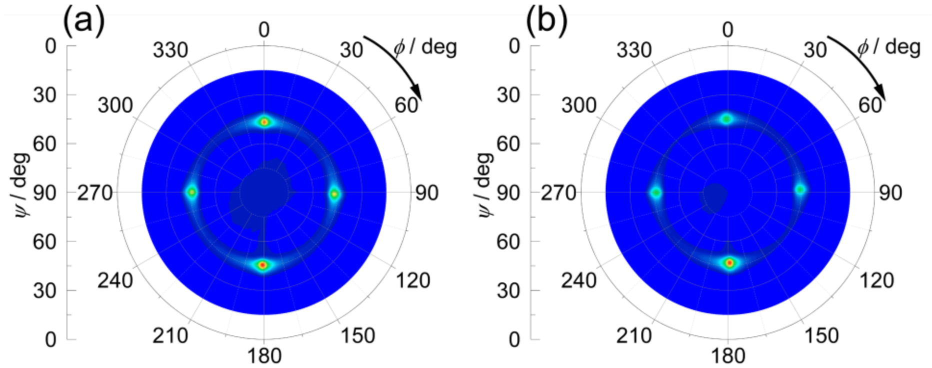

Figure 2. (103) pole figures of magnetically aligned Y123 powder samples with 10 T-MRF in (a) Resin A and (b) Resin B. Ψ and ϕ indicate tilt and rotation angles, respectively.

Figures 2(a) and 2(b) show (103) pole figures at α-plane for Y123 powder samples magnetically aligned under a 10 T-MRF in Resin A and Resin B, respectively. Our previous work [14] clarified that the *c*-axis was the first easy magnetization axis in Y123, and MRFs, it is expected to align perpendicular to the α-plane (see Fig. 1). Therefore, the α-plane is the most appropriate plane for measuring the (103) pole figure. In Fig. 2(a), sharp four-fold symmetric spots were observed at $\Psi \sim 45°$, resembling the (103) pole figure reported in our previous study [9]. These spots reflect the twin microstructures of Y123 grains, confirming their biaxial alignment under the 10 T-MRF in Resin A. Similarly, in Fig. 2(b), clear four-fold symmetric spots at $\Psi \sim 45°$ indicate that Y123 grains were also biaxially aligned under the 10 T-MRF in Resin B. The calculated biaxial orientation degrees were $F \sim 52$ % and $F \sim 50$ % for Resin A and Resin B, respectively. These results suggest that Y123 grains can achieve biaxial alignment under a 10 T-MRF regardless of the initial viscosity of the epoxy resin ($\eta_{init} \sim 0.5$ Pa·s vs. $\eta_{init} \sim 40$ Pa·s). This implies that at 10 T, the effects of η_{init} and t_{cure} are minimal due to the sufficient magnetic orientation energy and curing time.

Figures 3(a) and 3(b) show the (103) pole figures at the α-plane for Y123 powder samples magnetically aligned under a 1 T-MRF in Resin A and Resin B, respectively. In Fig. 3(a), the (103)

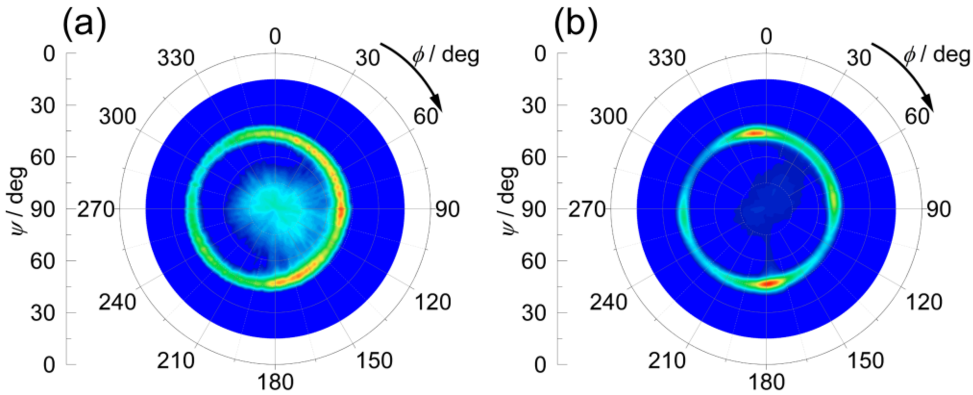

Figure 3. (103) pole figures of magnetically aligned Y123 powder samples with 1 T-MRF in (a) Resin A and (b) Resin B. Ψ and ϕ indicate the tilt and rotation angles, respectively.

pole figure exhibits a circular shape at $\Psi \sim 45°$, indicating that c-axis grain alignment was achieved in Resin A. In contrast, Fig. 3(b) shows the four-fold symmetric spots with the broad streaks along the ϕ direction at $\Psi \sim 45°$, suggesting that Y123 grains with twin microstructures were incompletely biaxially aligned in Resin B. The calculated biaxial orientation degrees were $F \sim$ 13 % and $F \sim$ 23 % for Resin A and Resin B, respectively. These results indicate that under a 1 T-MRF, biaxial orientation degrees can be enhanced by the effects of η_{init} and/or t_{cure}.

To understand the effects of η_{init} and t_{cure} on the biaxial orientation degrees in Y123, Figure 4 presents the relationship between F and B_{a} for Y123 powder samples aligned in Resin A and Resin B. Note that F includes contributions from both c-axis and in-plane orientation degrees. Focusing on the results for Resin A, the F value showed approximately 12 % at B_{a} = 0.8 T and was almost unchanged with the increase in B_{a} up to B_{a} = 5 T. However, at B_{a} = 10 T, F significantly improved to approximately 55 %, indicating a remarkable enhancement compared to the range B_{a} = 0.8 – 5 T. For Resin B, the F values at B_{a} = 0.8 T and 1 T were approximately 21 % and 23 %, respectively, showing no significant difference. However, at B_{a} = 3 T, F increased drastically to 42 %, and further improved to approximately 50 % at B_{a} = 10 T. A common trend observed in Fig. 4 is that the F values improve with increasing B_{a}, which can be qualitatively explained by the relationship between magnetic orientation energy and B_{a}. In general, the magnetic orientation energy is proportional to B_{a}^2, leading to enhanced biaxial orientation degrees as B_{a} increases.

Here, we compare the results for Resin A and Resin B in detail. The most obvious point in Fig. 4 is that the clear differences in the B_{a} dependence of F between Resin A and Resin B, except at B_{a} = 10 T. At B_{a} = 10 T, no clear differences in F were observed between the two resins. However, at B_{a} = 0.8 T, F for Resin B was higher than that for Resin A. Moreover, for B_{a} = 3 and 5 T, the differences in F between Resin A and Resin B was more pronounced compared to B_{a} = 0.8 T. From these experimental results, it was found that Y123 exhibited high biaxial orientation degrees at B_{a} = 10 T for $\eta_{\mathrm{init}} \sim$ 40 Pa·s, and at B_{a} > 3 T for $\eta_{\mathrm{init}} \sim$ 0.5 Pa·s. This indicates that F is influenced by the type of epoxy resin. Considering the specifications of Resin A and Resin B in Table 1, it is strongly suggested that lower η_{init} and/or longer t_{cure} contribute to higher biaxial orientation degrees in Y123.

As mentioned in Introduction, τ is proportional to η, and inversely proportional to $\Delta\chi$ and B_{a}^2. In principle, the magnetic alignment of grains should be completed before the resin fully cures, meaning τ should be equal to or shorter than t_{cure}. As shown in Table 1, η_{init} of Resin A is 80 times higher than that of Resin B, which implies that τ in Resin B is 80 times shorter than that in Resin A. In addition, t_{cure} of Resin B is approximately 1.7 times longer than that of Resin A. Based on this theoretical framework, the nearly constant F values observed at B_{a} = 10 T can be explained by the fact that τ remains significantly shorter than t_{cure} for both resins. In contrast, for 0.8 T < B_{a} < 5 T, the obvious differences in F were observed, likely due to the reversal in the relative magnitudes of τ and t_{cure} between Resin A and Resin B. Specifically, in Resin B, τ remains shorter than t_{cure}, facilitating alignment, whereas in Resin A, τ exceeds t_{cure}, limiting alignment. For Y123, which has a relatively low $\Delta\chi$, achieving sufficiently short τ is crucial under low B_{a} MRF conditions. A viable approach is selecting a dispersing medium with lower η_{init}. As observed in Fig. 4, Y123 showed higher F at B_{a} = 10 T for $\eta_{\mathrm{init}} \sim$ 40 Pa·s and at B_{a} > 3 T for $\eta_{\mathrm{init}} \sim$ 0.5 Pa·s. Extrapolating from these results, achieving higher F under lower B_{a} (e.g., at permanent magnet levels) is expected with a colloidal solution with η_{init} < 0.1 Pa·s. Since the colloidal process is a well-established ceramic fabrication technique, combining MRF at permanent magnet levels with a colloidal process is expected to be a promising method for producing magnetically biaxial aligned Y123 ceramics with high orientation degrees.

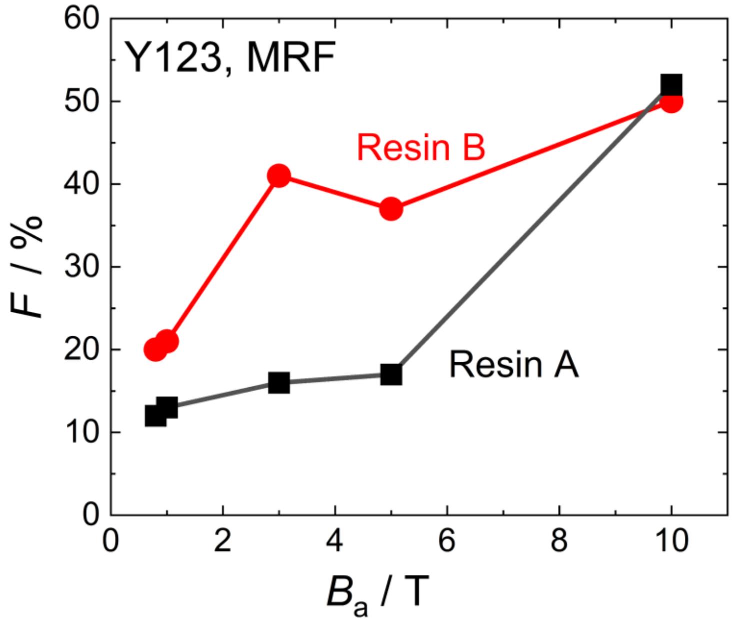

Figure 4. B_a dependence of F for the magnetically aligned Y123 powder samples with Resin A and Resin B. The applied B_a were 0.8, 1, 3, 5, and 10 T.

4. Conclusion

In this study, we investigated the biaxial orientation degrees of magnetically aligned Y123 powder samples using Resin A and Resin B. We successfully fabricated the biaxially aligned Y123 under a MRF at a permanent magnet level ($B_a \sim 1$ T) using Resin B, which has lower η_{init} and a longer t_{cure}. The biaxial orientation degrees for Y123, which has the smallest magnetic anisotropy among RE123 compounds, were strongly influenced by η_{init} and $t_{cure,}$ in the B_a region of 0.8 T < B_a < 5 T. Our findings suggest that the required B_a for effective magnetic alignment is closely related to the properties of the dispersing media, such as its initial viscosity and casting/curing time. To fabricate the RE-based cuprate superconducting materials using magnetic alignment technique, the combination of magnetic alignment with a colloidal processing approach is essential. Colloidal solution typically has $\eta_{init} < 0.1$ Pa·s, leading to significantly shorter τ. If an appropriate casting time is achieved, combining LDT-MRF with a permanent magnet array ($B_a \sim 1$ T) and a colloidal solution offers a practical route for producing magnetically biaxial aligned Y123 ceramics.

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