Engineering nanocolumnar defect configurations for optimized vortex pinning in high temperature superconducting nanocomposite wires

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We report microstructural design via control of BaZrO3 (BZO) defect density in high temperature superconducting (HTS) wires based on epitaxial YBa2Cu3O7-δ (YBCO) films to achieve the highest critical current density, $J_c$, at different fields, $H$. We find the occurrence of $J_c(H)$ cross-over between the films with 1–4 vol% BZO, indicating that optimal BZO doping is strongly field-dependent. The matching fields, $B_w$, estimated by the number density of BZO nanocolumns are matched to the field ranges for which 1–4 vol% BZO-doped films exhibit the highest $J_c(H)$. With incorporation of BZO defects with the controlled density, we fabricate 4-μm-thick single layer, YBCO + BZO nanocomposite film having the critical current ($I_c$) of ~1000 A cm⁻¹ at 77 K, self-field and the record minimum $I_c$, $I_c(\text{min})$, of 455 A cm⁻¹ at 65 K and 3 T for all field angles. This $I_c(\text{min})$ is the largest value ever reported from HTS films fabricated on metallic templates.

Synthesis of epitaxial nanocomposite films containing self-assembled, ordered arrays of nanodots/nanorods/nanopillars of one type of complex material embedded in another complex functional material has stimulated research in a wide range of application areas such as multiferroics, magnetoelectrics, thermoelectrics, ultra-high density storage, and high temperature superconductors¹–⁵. In these application areas, the nanocomposite films can offer new or significantly enhanced functionalities which are not attainable from individual materials or phases comprising the composite. One of the fascinating examples is high temperature superconducting (HTS) nanocomposites based on YBa2Cu3O7-δ (YBCO) containing a few volume% (vol%) insulating columnar defects comprised of simple or double perovskite-structured materials with simple (or distorted) cubic structure that include BaZrO3 (BZO), BaSnO3, Ba2RETaO6, and Ba2RENbO6⁵–¹². Driscoll et al.¹³ were the first to demonstrate that incorporation of randomly oriented, disk shaped BZO nanoparticles with a modal size of ~10 nm was beneficial to pinning¹³. It was subsequently demonstrated that spontaneous self-assembly of well-ordered, vertically aligned, nanocolumns parallel to the YBCO c-axis was possible by incorporation of the materials listed above¹³–¹² and that such self-assembly occurs via spontaneous phase separation and a strain-driven ordering process driven by minimization of strain fields arising from the lattice mismatch (5–12%) between the embedded nanophase and the YBCO matrix¹⁴. Since these columnar defects typically form ordered arrays of columns with diameters (5–10 nm) comparable to the diameter of a magnetic vortex core ($\sim 2 \times \xi(T)$, $\xi(T)$ = superconducting coherent length for YBCO, $\zeta^2(T)\approx\zeta^2(0)\left[1-\frac{T}{T_c}\right]$, $\zeta(0) = 1.5 \sim 2$ nm), they effectively pin or immobilize magnetic flux lines (or vortices), resulting in a dramatic improvement of the critical current density, $I_c$, in the presence of applied magnetic fields, particularly when the field is parallel to the orientation of the aligned defects, $H || c$-axis¹³–¹².

HTS wires based on YBCO nanocomposite films are an essential component to realize a wide range of environmentally friendly, highly efficient electric power devices with small weight and footprint¹⁵. Since the field and temperature regimes in which these devices are operated are different¹⁵, vortex pinning via columnar defects needs to be tailored to achieve the highest $I_c$ at the desired fields and temperatures required by the specific applications. An ideal strategy to achieve such a goal is to incorporate as many columnar defects as the number of vortices generated at the operating magnetic field. Fewer defects would leave unpinned vortices, and extra defects would compromise the volume of superconducting phase, both limiting $I_c$¹⁶. Since vortex spacing depends on the
magnetic field strength, nanocolumn spacing to accommodate all vortices is expected to be field-dependent. For this reason, the effect of BZO nanocolumn density on the \( I_c \) as a function of magnetic field strength, orientation, and operating temperature needs to be comprehensively understood. Ultimately, we should be able to tailor the BZO defect density to achieve an optimal superconducting performance at a given application specific, operating temperature and applied magnetic field. Although previous work has reported the importance of controlling the columnar defect density to enhance and tailor the in-field \( I_c \) properties\(^{15-25} \), there is no report of systematic variation of the optimal columnar defect density as a function of magnetic field strength, orientation, and operating temperature.

In the present work, we show that the density of BZO columnar defects necessary to achieve optimal \( I_c \) performance systematically increases with increasing magnetic field and is also influenced by field orientation and temperature. In addition, we demonstrate that a single layer 4-\( \mu \)m-thick YBCO films with the controlled density of BZO nanocolumns exhibits excellent critical current (\( I_c \)) performance given by a \( I_c \) of \( \sim 1000 \) A cm\(^{-1} \) at 77 K, self-field and a minimum \( I_c, I_c(\text{min}) \), of 455 A cm\(^{-1} \) at 65 K and 3 T for all magnetic field orientations. Such high \( I_c(\text{min}) \) at 65 K and 3 T is the record value ever reported from HTS films fabricated on metallic templates.

**Results**

**Engineering BZO nanocolumnar defect density for \( I_c \) optimization at different fields.** Figure 1 summarizes X-ray diffraction results for YBCO films with different BZO contents up to 10 vol\%. All samples with same thickness of \( \sim 0.8 \) \( \mu \)m were deposited on LMO/IBAD-MgO/Hastelloy templates via pulsed laser deposition (PLD). The detailed procedures of the samples preparation are summarized in the Experimental section. The 0–20 scans (Fig. 1a) show that all films have sharp out-of-plane c-axis orientation with strong (00f)

peak intensities of YBCO phase. For BZO-doped samples, additional peaks at \( 2\theta = 21.6^\circ \) (which is evident only for the 10 vol\% BZO-doped sample due to the large concentration of BZO phase) and 43.3\(^\circ \) correspond to (001) and (002) BZO peaks indicating the formation of epitaxial BZO nanophase within the YBCO matrix. The narrow scans (Fig. 1b) for BZO (002) peak measured at the maximum X-ray power show evidently stronger peak intensities with increasing BZO addition, despite the slight peak overlap with the MgO (200) peak, which stems from the epitaxial MgO layer comprising the template. Detailed XRD analysis based on peak broadening estimates that the size of BZO nanophase is \( \sim 6–7 \) nm and remains nearly unchanged up to 10 vol\% BZO. Figure 1c shows the summary of full-width-half-maximum of omega and phi scans (\( \Delta \omega \) and \( \Delta \phi \)) for YBCO and BZO phases taken from the samples. Essentially, no meaningful variation of \( \Delta \omega \) and \( \Delta \phi \) in both YBCO phase and BZO nanophase is measured from all BZO-doped films. The \( \Delta \omega \) and \( \Delta \phi \) values of YBCO phase are in the ranges of 1.3\(^\circ \) to 2\(^\circ \) and 3.2\(^\circ \) to 3.6\(^\circ \), respectively, while those of the BZO nanophase range from 2.1\(^\circ \) to 2.8\(^\circ \) and from 5.8\(^\circ \) to 6.2\(^\circ \), respectively. Relatively larger \( \Delta \omega \) and \( \Delta \phi \) for the BZO nanophase should be attributed to the deviation of their alignments with respect to the in-plane and out-of-plane of YBCO film as well as the substrate\(^{10} \). Although the \( \omega \) and \( \phi \) scan results imply that all samples with BZO contents up to 10 vol\% have good epitaxial YBCO phase with similar in-plane and out-of-plane textures, YBCO (113) pole figure measurements (Fig. 1d) reveal that texture degradation occurs when BZO content increases to 10 vol\%. Unlike the 1–4 vol\% BZO-doped samples having only strong, sharp four-fold symmetric peaks, the 10 vol\% BZO-doped film exhibits an additional texture component indicated by a circular pattern along with the four-fold symmetric peaks, mostly caused by formation of misoriented and 45\(^\circ \) rotated grains. A quantitative fraction (%cube-texture) of “cube-textured”

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**Figure 1** | X-ray diffraction results for YBCO films with different BZO additions. (a) 0–20 scans for the samples at X-ray power (–50 kV, 5 mA) where the intensity of (006) YBCO peak does not exceed the maximum intensity limit of the detector. (b) 0–20 scans from \( 2\theta = 41.5^\circ \) to 45.5\(^\circ \) at maximum X-ray power (-50 kV, 30 mA). (c) \( \Delta \omega \) and \( \Delta \phi \) values for YBCO phase and BZO nanophase for the films with and without BZO additions. (d) YBCO (113) pole figures for 1–10 vol\% BZO-doped samples.
volume out of the total volume of the layer was determined by the numerical method described elsewhere. The 10 vol% BZO doped sample only has 90% cube-texture which is smaller than the values of 95 ~ 96% typically measured from un-doped and 1~4 vol% BZO-doped films.

Transmission electron microscopy (TEM) examination confirms the formation of self-aligned BZO nanocolumns in YBCO + BZO nanocomposite films. As shown in Figs. 2a and 2c, cross-section TEM images for the 1 and 4 vol% BZO-doped films show BZO nanocolumns, well-ordered and in general aligned to the crystallographic c-axis of YBCO with some splayed feature with respect to the c-axis similar to previous reports. Figure 2b shows a plan-view TEM image for the 2 vol% BZO-doped film containing moiré fringes caused by BZO nanoparticles which are sections of BZO nanocolumns. The TEM images confirm the increase in the columnar defect density with BZO content while their average diameters remain unchanged around 5~6 nm, consistent with the estimated values by XRD. With the nanocolumn spacing, we calculated the matching field, $B_w = \frac{2\Phi_0}{3\pi a^2}$ as ~0.8 T for 1 vol% BZO, ~6 T for 2 vol% BZO, and ~15 T for 4% BZO-doped samples, where $\Phi_0 = 2.07 \times 10^{-15}$ T cm$^2$ is the flux quantum and $a$ is the vortex spacing that matches $d$ at $B_w$.

When BZO doping is further increased to 10 vol%, we observed a morphological change in the array of BZO defects as shown in Fig. 3. Essentially, some BZO defects start to be aligned horizontally (along $ab$-planes) along with BZO nanocolumns aligned vertically (along $c$-axis) denoted by blue and red arrows (Fig. 3a). Since the imaging conditions that bring out BZO defects aligned vertically and horizontally are slightly different, the defects aligned to the $ab$-planes are more clearly observed in a different image of the same specimen taken from different imaging conditions (Fig. 3b). Vertically aligned BZO nanocolumns are very dense, and the average BZO column spacing can be estimated to be less than 10 nm, much narrower than that for the 4 vol% BZO-doped sample. The appearance of semi-continuous, planar BZO defect arrays is anticipated when the BZO doping concentration is high enough to minimize the distance between adjacent BZO nanodots nucleating within the same $ab$-plane, as shown in our previous work. Further increase in BZO content is expected to produce continuous planar BZO layers in the YBCO matrix. Indeed, YBCO/BZO multilayer structure containing planar BZO defects arrays was reported from YBCO + BZO nanocomposite films with high BZO concentration such as 50 mol% YBCO and 50 mol% BZO.

Table 1 summarizes superconducting transition temperature, $T_c$, and self-field $J_c$ at 77 K and 65 K for the set of un-doped and BZO-doped YBCO films with different BZO contents. Note that as denoted in Table 1, 1 vol% BZO is equivalent to 1 wt% and 2.4 mol% BZO. Several 0.5 and 1 vol% BZO-doped films are reproducibly measured to have $T_c$ of 87.5 ~ 87.7 K, essentially unchanged from those ($T_c = 87.5 ~ 88.0$ K) obtained from pure YBCO films grown on IBAD-MgO templates in identical PLD conditions. When BZO doping is over 1 vol%, $T_c$ starts to drop monotonically with a drop rate of ~0.46 K/vol% (0.2 K/mol%). In a previous work, we suggested that the origin of $T_c$ reduction with increasing BZO doping should be attributed to local strain field caused by the lattice mismatch (~8%) between YBCO and BZO phases and the consequent local oxygen deficiency that possibly occurs around BZO nanocolumns. Microstrain measurement of BZO-doped samples based on Williamson-Hall plots shows that the strain field monotonically
increases as BZO content increases. High resolution, scanning TEM analysis confirms that a local area surrounding a BZO nanocolumn having approximately an outer diameter of ~15–20 nm, is highly strained and also oxygen deficient. When average BZO columns spacing is near or above this diameter by increasing BZO doping, an overlap of strained REBCO regions surrounding BZO nanocolumns is expected to occur, and $T_c$ starts to decrease. As aforementioned, the BZO doping level that results in the average BZO column spacing near or above ~15–20 nm is around 2–4 vol%, which is the concentration at which $T_c$ apparently starts to decrease. For the self-field $J_c$ at 77 K and 65 K, the $J_c$ values are noticeably improved with BZO doping up to 1 vol%. The highest $J_c$ of 4.1 MA cm$^{-2}$ at 77 K and 12.2 MA cm$^{-2}$ at 65 K are achieved from the 1 vol% BZO-doped sample. After an initial enhancement, the $J_c$ gradually degrades up to 0.34 MA cm$^{-2}$ at 77 K with further increasing BZO up to 10 vol%. Such $J_c$ variation suggests that even at self-field, additional BZO defects enhance the pinning and $J_c$ performance of the films, and 1 vol% BZO is an optimum doping level resulting in the highest $J_c$ at 77 K and 65 K.

In the presence of applied magnetic fields, the strong dependence of the optimum BZO doping level on the applied magnetic field strength is observed. Figure 4 shows $J_c$ and pining force ($F_p$) as a function of applied magnetic field, $J_c(H)$ and $F_p(H)$, for YBCO films with different BZO contents, for the field parallel to YBCO c-axis, $(H\parallel c)$ at 77 K and 65 K. The inset of Fig. 4a also displays the normalized $J_c(H)$ at 77 K to determine $\alpha$ values in the power-law dependence of $J_c$ on $H$, $J_c \sim H^{-\alpha}$ valid in the range of ~0.2–1.5 T, which are also summarized in Table 1. Compared to the undoped sample, the 1–4 vol% BZO-doped samples show a significantly improved $J_c(H)$, much higher $F_p(H)$, and smaller $\alpha$ values, indicating the efficacy of BZO columnar defects as strong c-axis pinning centers. The highest $F_p$ (max) values of 9.5 GN m$^{-3}$ at 77 K, 2 T and 74 GN m$^{-3}$ at 65 K, 5 T as well as the smallest $\alpha$ value of ~0.33 are obtained from the 2 vol% BZO-doped sample. Unlike these samples, 10 vol% BZO-doped sample shows much poorer $J_c(H)$ and $F_p(H)$ at both temperatures, primarily due to the detrimental problems caused by excess BZO doping as will be discussed later. Another essential feature observed in the figures is the occurrence of $J_c(H)$ (or $F_p(H)$) cross-over between the films with 1–4 vol% BZO, indicating that the optimal BZO doping is strongly field-dependent. The $J_c(H)$ data at 77 K show that the 1 vol% BZO-doped film has the highest $J_c(H)$ in the self-field and up to the first cross-over field of ~1.6 T. When the field is higher than ~1.6 T, the 2 and 4 vol% BZO doping levels lead to the highest $J_c(H)$ up to and over the second cross-over field of ~7.3 T, respectively. $J_c(H)$ data at 65 K also show the same trend, but the cross-over fields are moved to higher fields as indicated in the figure.

Figures 5a–5c show angular dependent $J_c$, $I_c(\theta)$, for the samples at different temperatures and field regimes. The measurement was made in the maximum Lorentz force configuration in which the applied field direction is always perpendicular to the direction of current flow. 1–4 vol% BZO-doped films show higher $J_c(\theta)$ than the undoped sample for the entire angular range. In addition to higher $J_c(\theta)$ at and near $H\parallel c$ stemming directly from strong c-axis correlated pinning via BZO nanocolumnar defects, the overall improvement in $J_c(\theta)$ for angles away from $H\parallel c$ can be attributed to the enhanced pinning via strong BZO defect-vortex interaction that still exist, although the pinning strength gradually decreases due to the reduced area of vortex confined by BZO columns with angles away from $H\parallel c$. This is the primary reason why the enhancement in $J_c(\theta)$ near $H\parallel ab$ for 2 and 4 vol% BZO-doped samples is limited and eventually negligible at 65 K, 3 and 5 T. Despite this fact, the 1 vol% BZO-doped sample maintains higher $J_c(\theta)$ near $H\parallel ab$ than the 2 and 4 vol% BZO-doped samples, which lead to the highest $J_c(\theta)$ for all field orientations, as shown in Fig. 5d. Note that the $J_c(\theta)$ is considered a critical performance parameter particularly for devices with wound wire architectures such as motors, generators, and high field magnets, so that HTS wires having higher $J_c(\theta)$ are more desirable for these device applications.

| Table 1 | Transport $T_c$, self-field $J_c$ at 77 K and 65 K, and $\alpha$ values for 0.8 μm thick YBCO + BZO nanocomposite films grown on IBAD-MgO templates via PLD |
| YBCO vol.% BZO | $T_c$ (K) | $J_c$ (77 K, sf) (MA cm$^{-2}$) | $J_c$ (65 K, sf) (MA cm$^{-2}$) | $\alpha$, $J_c(H^{-\alpha})$ |
| Pure YBCO | 0 | 87.7 | 2.8 | 7.6 | 0.48 |
| 0.5 vol% BZO | 1.2 | 87.5 | 3.2 | 8.1 | 0.41 |
| 1 vol% BZO | 2.4 | 87.7 | 4.1 | 12.2 | 0.33 |
| 2 vol% BZO | 4.8 | 86.6 | 3.0 | 8.6 | 0.33 |
| 4 vol% BZO | 9.6 | 84.9 | 2.4 | 7.4 | 0.42 |
| 10 vol% BZO | 24 | 83.6 | 0.34 | 2.2 | 0.57 |

Growth of thick YBCO films with engineered nanocolumns. Considering $J_c(H,\theta,T)$ data for ~0.8 μm thick YBCO nanocomposite films with different BZO contents reported in Figs. 4 and 5, we grew ~4-μm-thick YBCO films containing 1 vol% BZO in order to achieve superior $J_c$ performance at self-field and in...
Figure 4 | Field dependent $J_c$ and $F_p$ for $H//c$ with the magnetic field up to 8 T for pure YBCO and YBCO + 1–10 vol% BZO films. (a) $J_c(H)$ at 77 K, (b) $J_c(H)$ at 65 K, (c) $F_p(H)$ at 77 K, and (d) $F_p(H)$ at 65 K. The inset of Fig. 4(a) shows normalized $J_c$, $J_c(H)/J_c(sf)$, for the samples. All samples have an identical film thickness of ~0.8 μm.

Figure 5 | The angular dependent $J_c$ at (a) 77 K, 1 T, (b) 65 K, 3 T, and (c) 65 K, 5 T for pure YBCO and YBCO + 1–10 vol% BZO films, in magnetic field applied −30° to 110° with respect to the $c$-axis of the film. (d) $J_c$ (min) for all field orientation for YBCO nanocomposite films with different BZO additions.
moderate magnetic field range of 0.5–3 T at 77 K and 65 K. To quantitatively evaluate the performance improvement by incorporating BZO defects, 4-μm-thick, un-doped films were also prepared in identical growth conditions. Phase and microstructural properties for both 4-μm-thick HTS films with and without 1 vol% BZO are summarized in Fig. 6. The most striking difference in structural properties between the un-doped and BZO-doped samples is the amount of a-axis oriented grains which are one of the major detrimental factors precluding a proportional increase of \( I_c \) with thickness\(^{26}\). Quantitative volumetric amount of a-axis grains in the films is determined by XRD, comparing the intensity of (001) normal with (100) normal peaks obtained in YBCO (102) \( \chi \)-scans, \( I_{(001)}/[I_{(001)}+I_{(100)}] \times 100 \), as shown in Fig. 6b. The BZO-doped thick film is measured to have only \( \sim 3 \) vol% of a-axis oriented grains which is remarkably reduced from \( \sim 30 \) vol% of a-axis oriented grains measured from the un-doped thick sample. Plan-view scanning electron microscopy images (Figs. 6c and 6d) show that the BZO-doped thick film has denser and smoother surface morphology with significantly reduced a-axis oriented surface particles/grains compared to the un-doped thick film.

Figure 7 shows transport \( I_c \) versus magnetic field at 77 K and 65 K for 4-μm-thick films with and without 1 vol% BZO. The \( I_c \) data at 75.5 K for the film containing 1 vol% BZO were also measured and shown in the inset of Fig. 7b for comparison with the previously reported high \( I_c \) data at 77 K\(^{27,28}\). The BZO-doped thick film has the self-field \( I_c \) values of 995 A cm\(^{-1}\) at 77 K and 1188 A cm\(^{-1}\) at 75.5 K, which correspond to the \( J_c \) of 2.5 and 3 MA cm\(^{-2}\), respectively. The self-field \( I_c \) at 77 K for the BZO-doped sample is more than twice as large as the \( I_c \) of 465 A cm\(^{-1}\) of the un-doped thick film. In fact, multiple samples containing 1 vol% BZO showed the self-field \( I_c \) of 750–1000 A cm\(^{-1}\) at 77 K, while un-doped samples have the \( I_c \) of 450–530 A cm\(^{-1}\). Due to a limit in the maximum current (~25 A) that can be applied to the patterned sample (215 μm in width), the self-field \( I_c \) at 65 K cannot be directly measured. However, the \( I_c \) over 1000 A cm\(^{-1}\) at 65 K are retained at fields up to 1 T for \( H//c \) and 1.5 T for \( H//ab \). In the presence of applied magnetic fields, superior \( J_c(H) \) with excellent \( I_c \) retention properties can be seen in the BZO-doped thick sample, compared to the un-doped thick film. At 77 K, 1 T, the \( I_c \) of \( \sim 340 \) A cm\(^{-1}\) for \( H//c \) is only reduced by a factor of \( \sim 2.9 \) from the \( I_c \) at self-field, indicating a massive enhancement in pinning via BZO columnar defects. At 65 K, the \( I_c \) for \( H//c \) and \( H//ab \) are well over 150 A cm\(^{-1}\) at fields up to 8 T. All these \( I_c \) values are 2- or 3-fold larger than those for the un-doped thick sample.

Angular dependent \( I_c \) at 77 K, 1 T and 65 K, 3 T are shown in Fig. 8. Compared to the un-doped thick film, the BZO-doped thick film shows the remarkably improved \( I_c \) for all field angles. The BZO-doped thick film has the \( I_c\)(min) of 196 A cm\(^{-1}\) at 77 K, 1 T and 455 A cm\(^{-1}\) at 65 K, 3 T which are about three times larger than those for the un-doped thick film. At 75.5 K, 1 T, the BZO-doped film is also measured to have the \( I_c\)(min) of 251 A cm\(^{-1}\) (the data not shown here) which is nearly 30% increase in the value at 77 K, 1 T.

Discussion

Since \( B_w \) is the field at which the density of vortices matches that of the BZO nanocolumns, Figure 2d provides key information on how
much BZO doping is approximately required to accommodate vortices generated in a specific field range\(^9\). For instance, 1 and 2 vol% BZO should be the optimum levels to maximize the pinning and \(I_c\) performance at low fields of \(\sim 0.8\) T and modest fields of \(\sim 6\) T respectively, but the doping level should increase to 4 vol% at much higher fields around 15 T. 10 vol% BZO-doped samples contain very dense BZO defects aligned both parallel and perpendicular to the YBCO c-axis that may effectively enhance the pinning for fields over 15 T (see Fig. 3). However, significant reduction in YBCO phase volume fraction (proportional to the amount of BZO volume fraction) as well as the degradation of cube texture (see Fig. 1d and Table 1) via excess BZO doping should result in negative impacts on the \(I_c\) particularly at high temperatures of 77 K and 65 K, over any gain caused by the enhanced pinning via the BZO defects as shown in Figs. 4 and 5. When comparing the field ranges for which the 1–4 vol% BZO-doped films exhibit the highest \(I_c(H,T)\) in Fig. 4 with the \(B_g\) values estimated from the respective BZO column densities, we find that the \(B_g\) values are close to or within these field ranges. The results demonstrate that optimal BZO nanocolumns density to achieve the highest \(I_c\) is different at different fields and temperatures, dictating that the number density of columnar defects is an effective way to achieve the highest \(I_c\) at any targeted field-temperature range.

Understanding of the effect of BZO doping on \(I_c(H,\theta)\) at 77 K and 65 K for YBCO + BZO nanocomposite films provides key information to achieve high \(I_c\) performance at technically important field-temperature ranges for various electric power applications, such as i) 0–0.5 T, 65–77 K (for underground transmission cable, transformer, and fault current limiter) and ii) 3–5 T, 65 K (for large-scale motors and generators)\(^{15}\). Growth of a single layer YBCO film with high \(I_c\) (77 K, sf) over 1000 A cm\(^{-2}\) has been considered a very challenging work due to an exponential decay of critical current density, \(J_c\), making a proportional increase in \(I_c\) with the film thickness not feasible. The decay in \(I_c\) with thickness can be attributed to a number of factors including roughening of film, loss of epitaxy or degradation of texture and reduction in the defect density of naturally formed defects\(^{26,29,30}\). To date, there have been only a few reports on fabrication of a single layer, thick YBCO films having self-field \(I_c\) close to or over 1000 A cm\(^{-2}\) at 75.5–77 K\(^{27,28,31}\). Very recently, \(I_c\) (77 K, sf) of 1018 A cm\(^{-2}\) was reported in 5.9 \(\mu\)m thick DyB\(_2\)Cu\(_3\)O\(_7\)-s grown on the metallic templates prepared by inclined substrate deposition\(^{32}\). For the films grown on IBAD metallic templates, the self-field \(I_c\) of 944 A cm\(^{-2}\) at 77 K and 1007 A cm\(^{-2}\) at 76.55 K were obtained from a piece of the wire with 3.3 \(\mu\)m thick, 5 mol% Zr-doped (Y,Gd)BCO film\(^{16}\). For the films grown on single crystal substrates, the highest \(I_c\) of \(\sim 1500\) A cm\(^{-2}\) at 75.5 K, self-field was reported from 9 \(\mu\)m thick YBCO films containing 5 mol% BZO and 5 mol% Y\(_2\)O\(_3\)\(^{36}\). Recent reports suggest that incorporation of BZO and (or) other defects into thick YBCO films is a very effective means to achieve the improved \(I_c\) at self-field as well as in magnetic fields\(^{27,28,31}\).

Figure 9 compares the \(I_c\) and engineering critical current density, \(J_{c \text{E}}\), data at a field-temperature regime of 65 K, 3 T for the BZO-doped sample with those for the previous record performance HTS wires comprised of \(\sim 3\) and 4 \(\mu\)m thick YBCO + BZO nanocomposite films\(^{37}\). The \(I_{c \text{min}}\) (min) of 455 A cm\(^{-2}\) for the thick sample reported in this study corresponds to \(J_{c \text{E}}\) of 43.7 kA cm\(^{-2}\) and 84.2 kA cm\(^{-2}\) with and without considering a 50 \(\mu\)m thick Cu stabilizer which may be needed in application. Both \(I_c\) and \(J_{c \text{E}}\) performances are much higher than the previous values and are significantly exceeding the performance levels of 20 kA cm\(^{-2}\) needed for the applications\(^{38}\).
Higher $I_c$ and $J_c$ can provide wider gap from the operating current levels of a given application and thus, are always highly desirable as a greater safety buffer, to further reduce AC losses, and to avoid exceeding $I_c$, locally due to any perturbation.

In summary, we systematically investigated the influence of BZO nanocolumnar defect density on structural and superconducting properties of YBCO + BZO nanocomposite films. The nanocomposite films with up to 4 vol% BZO content have highly cube-textured, epitaxial YBCO phase and BZO nanophase with the morphology of vertically ordered nanocolumns. Compared to the undoped sample, the 1–4 vol% BZO-doped samples showed a significantly improved $I_c$ performance for entire magnetic field ranges with much higher $F_p$ and smaller $\alpha$ values due to the presence of strong pinning via BZO columnar defects. More importantly, we show that the optimum density of BZO nanocolumnar defects is strongly field-dependent and thus, different BZO doping levels are needed to optimize pinning and to achieve the highest $I_c$ properties at different magnetic fields, particularly when the field is parallel to the orientation of BZO defects, $H \parallel c$. Finally, we have fabricated thick, single-layer nanocomposite films with the controlled BZO concentration showing a self-field $I_c$ of ~1000 A cm$^{-2}$ at 77 K and the record $J_c$(min) of 455 A cm$^{-2}$ at 65 K and 3 T ever reported from HTS films fabricated on metallic templates.

**Methods**

All YBCO films with and without different vol% of BZO were prepared by pulsed laser deposition (PLD) using a KrF ($\lambda = 248$ nm) excimer laser at a repetition rate of 10 Hz. Laser energy density and substrate to target distance were optimized to be 2 J cm$^{-2}$ and 5 cm, respectively. The PLD target (2 inch diameter, 0.25 inch thick) was made by mixing commercially available YBCO powder and BaZrO$_3$ nanopowder using standard ball milling, followed by densification at 950°C for 2 h. The film growth temperature, $T_g$ was 790°C and the oxygen partial pressure, $P(O_2)$ was 230 mTorr. All depositions were performed on IBAD-MgO templates with a LaMnO$_3$ cap layer that were supplied from Supervapor Inc.

After deposition, samples were in-situ annealed at $T_a = 500$°C and $P(O_2) = 300$ Torr, and ex-situ annealed at 500°C for 1 h in flowing O$_2$ gas after depositing sputtered Ag electrodes onto the films. The 0.5 cm wide $\times$ 2 cm long sample was patterned into a 0.2 mm wide bridge by laser-scribing, due to the limits on the maximum measuring current in the characterization system. The standard four-point probe method with a voltage criterion of 1 $\mu$V cm$^{-1}$ was used for the transport measurements. Phase and textures of the films were characterized by x-ray diffraction method. Microstructures were characterized by transmission electron microscopy using a Hitachi HF-3300 TEM/STEM with a secondary detector and an energy-dispersive x-ray spectrometer at 300 kV and field emission scanning electron microscopy using Hitachi S4800 FE-SEM. TEM foils were prepared by the focus ion beam (FIB) technique, followed by low voltage ion milling and plasma cleaning.

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**Author contributions**

S.H.W. designed experiment, prepared the samples by PLD, characterized structural properties of the samples by XRD and SEM, and wrote the manuscript. Y.L.Z. measured superconducting properties of the samples. C.C. carried out the TEM characterization of the samples. A.G. supervised the research work and discussed the results. All authors reviewed the manuscript.

**Additional information**

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