Supplementary Material:

X-Ray Diffraction (XRD) of HfO₂ Nanocrystals.

Figure 1. XRD pattern of HfO₂ nanocrystals showing only reflections of the monoclinic phase after the heating-up synthesis.

Jetting and non-jetting waveforms of pristine and HfO₂-YBCO inks.

Figure 2. Optimized jetting and non-jetting waveforms used for printing (A) pristine and (B) HfO₂-YBCO inks. These waveforms were sequentially applied based on the pattern to be printed. Pristine YBCO inks were printed reproducibly with a print frequency of 12 kHz using the custom waveform with the ejection voltage applied to the piezoelectric nozzle in the range of 17–18 V for the jetting waveform and 9–10 V for the non-jetting waveform. The firing voltage of the piezoelectric nozzles was increased to 18–19 V with a lower print frequency of 9 kHz for HfO₂-YBCO inks.

Cross-Sectional Scanning Electron Microscopy (SEM) View of a Pristine and a HfO₂-YBCO Film Obtained via Single Ink-Jet Printing Deposition
Figure 3. Cross-sectional SEM image of YBCO (A) without and (B) with 5 mol% HfO₂ nanocrystals in one single deposition after full thermal process. Secondary phases like Ba₃Cu₂O₆ are marked with dotted lines.

Characterization of Pristine YBCO Films after Multi-Deposition

Figure 4. (A) Cross-sectional SEM view and (B) corresponding XRD spectrum of YBCO layer obtained via 5 times of multi-deposition, indicating presence of Ba(O,F)₂ phase above the YBCO layer.
Figure 5. Composition and microstructure after full thermal processing of an ink-jet-printed YBCO film with 4 deposition runs. (A) XRD analysis (asterisk: reflections by the secondary radiation of x-ray tube), (B) topographical SEM image, (C) cross-sectional SEM view of LaAlO3/YBCO architecture.

Transmission electron microscopy (TEM) analysis

Figure 6. (h00) bright-field TEM showing twin boundaries of (A) pristine and (B) HfO2-YBCO layer. The density of twin boundaries nearly doubled by the addition of HfO nanocrystals.