

## Supporting Information

### Graphene oxide as a template for a complex functional oxide

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#### Experimental section:

*Synthesis:* Graphene oxide was synthesized *via* the modified Hummers' Method.<sup>[1]</sup> The GO foam was prepared by a uni-directional freezing method, as reported previously.<sup>[2]</sup> GO papers were prepared by filtering a GO solution (1 mg mL<sup>-1</sup>) through an anodisc 0.2µm filter and cutting to the desired size.<sup>[1]</sup> The resulting monoliths were soaked for 48 h in a stoichiometric aqueous solution of yttrium, barium and copper nitrates (all Sigma Aldrich, UK) in a ratio of 0.05:0.1:0.15 M. The monoliths were then removed from the precursor solution and dried for 12 h at 60 °C, followed by calcination in an alumina boat-shaped crucible at 920 °C for 2 h, with a ramp rate of 10 °C min<sup>-1</sup>, and cooled to room temperature at a rate of 2 °C min<sup>-1</sup>. All calcinations were performed in an ELF 11/6 chamber furnace (Carbolite, UK) in air and at atmospheric pressure.

*Crystallochemical characterisation:* The phases present in the resulting monoliths of YBCO were analysed using powder X-ray diffraction (PXRD) (D8 Advance, Bruker, Germany) using Cu Kα radiation. The macro-scale structure was determined by scanning electron microscopy (SEM) (JSM 6330F, JEOL, Japan). High-resolution transmission electron microscopy (HRTEM) (JEM 2010, JEOL, Japan) was used to image the microscopic crystalline structure of the crystallites and selected area electron diffraction was used to determine the phases of the crystallites observed. Elemental content was determined using energy dispersive X-ray analysis (EDXA) (Oxford Cryosystems, UK) fitted to both the HRTEM and SEM. X-ray µ-computed tomography (NIKON XT H-225ST with a microfocus X-ray source with a 2,000 x 2,000 pixel Perkin Elmer 1620 detector) was used to determine the internal structure of the monoliths, with images produced using Avizo Fire software.

*Physical characterisation:* Magnetometry was performed using a superconducting interference device (SQUID) (Magnetic Property Measurement System Quantum Design, USA). Samples measured were used as synthesized and placed inside a gelatine capsule. Calculation of  $J_c$  was performed using the Bean critical state model.<sup>[3]</sup> In both cases the crystallite size was found from the measurement of SEM images, taking the smallest dimension, with the value for the foam being  $530 \pm 300$  nm and for the tape,  $460 \pm 400$  nm.

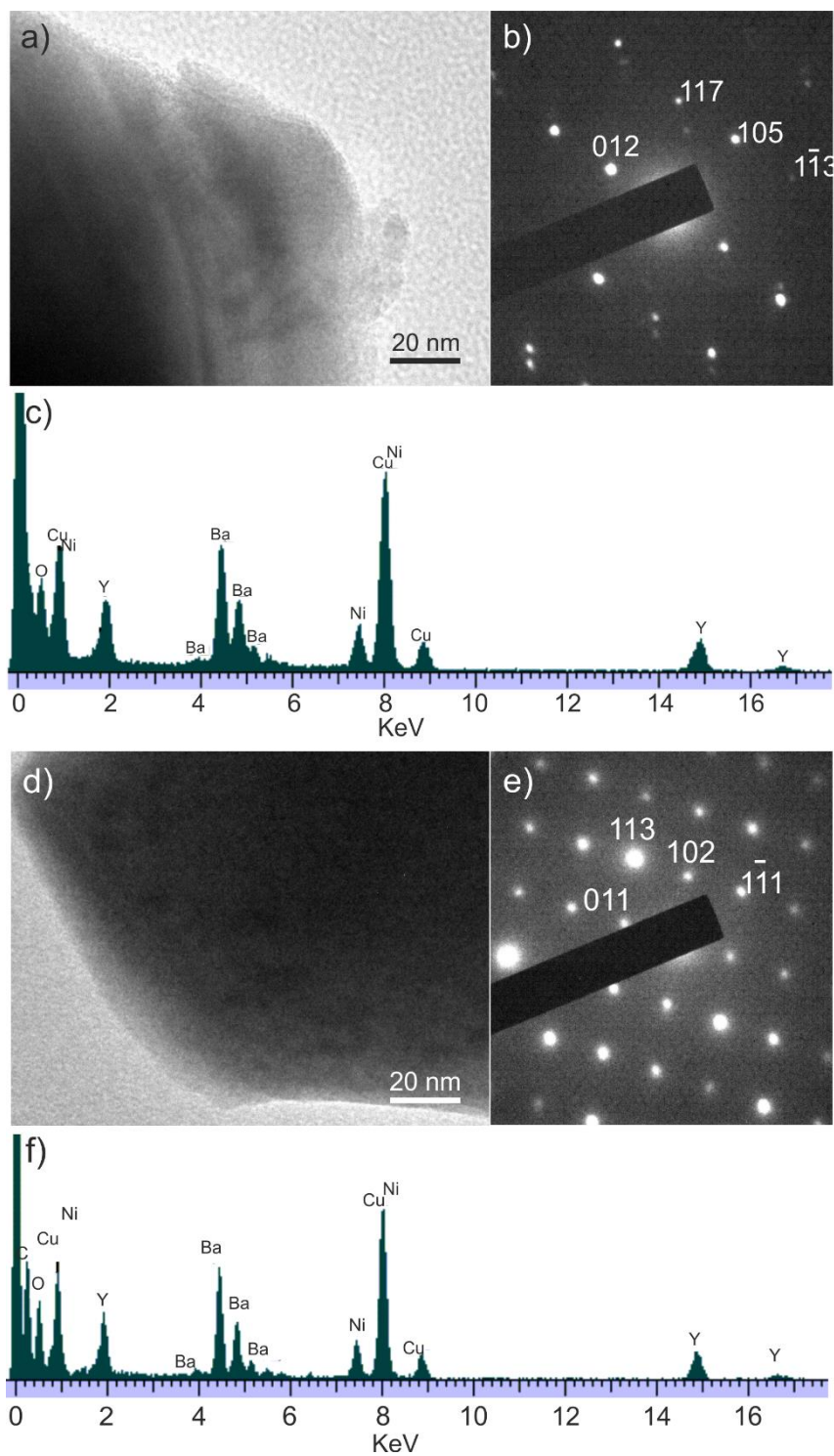


Figure S1. TEM micrographs of crystallites in a) the Y123 foam, with b) corresponding selected area diffraction indexed to the  $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$  phase, and c) corresponding EDXA. d) shows a crystallite of the Y123 tape with e) corresponding selected area diffraction indexed to the  $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$  phase, and f) corresponding EDXA. Nickel is present as the grid material

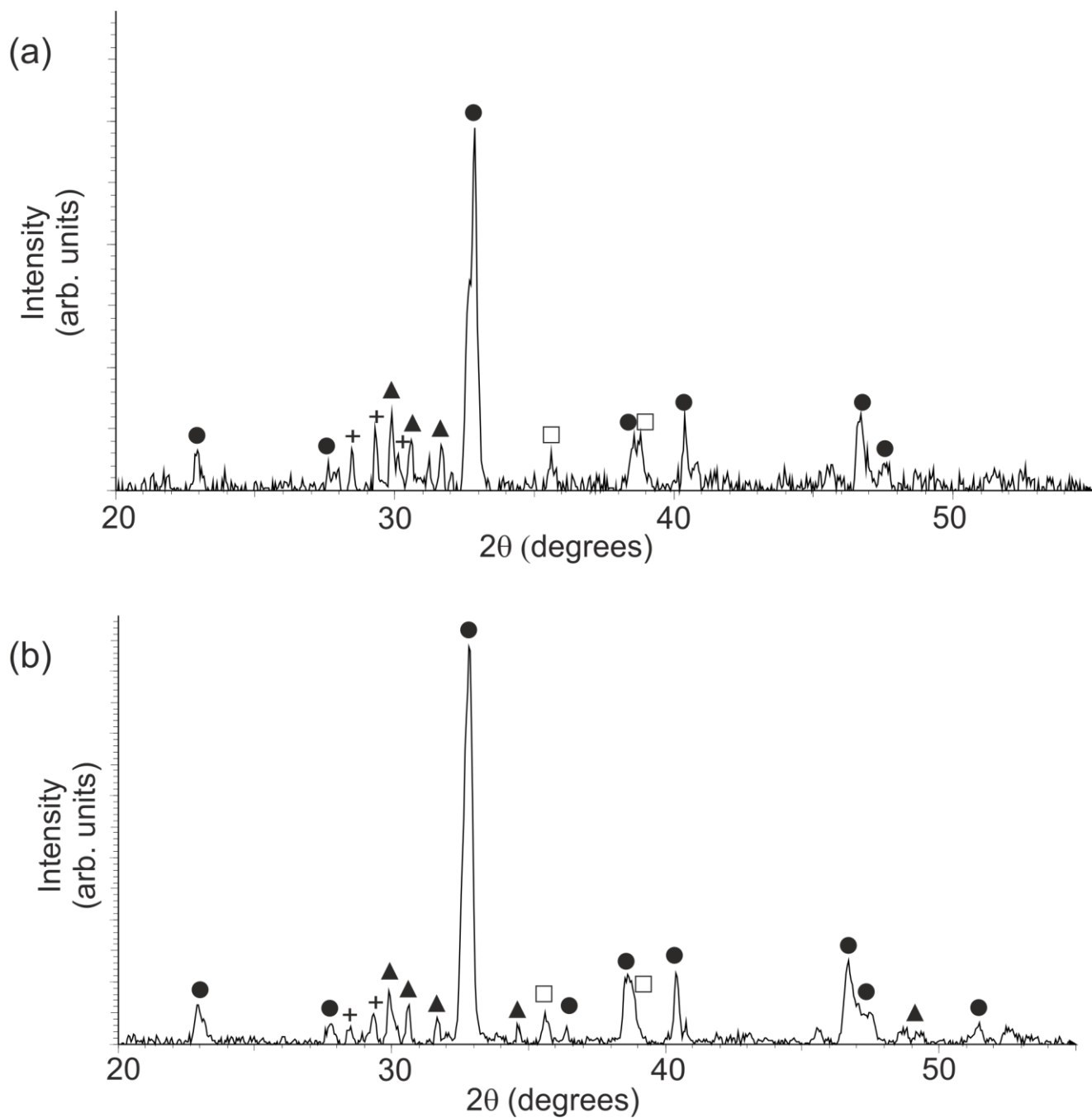


Figure S2. PXRD illustrating the crystalline phases present in a) the Y123 foam and b) the Y123 paper. Phases indexed are  $\text{YBa}_2\text{Cu}_3\text{O}_{6.9}$  (●),  $\text{CuO}$  (□),  $\text{Y}_2\text{BaCuO}_5$  (▲) and  $\text{BaCuO}_2$  (+).

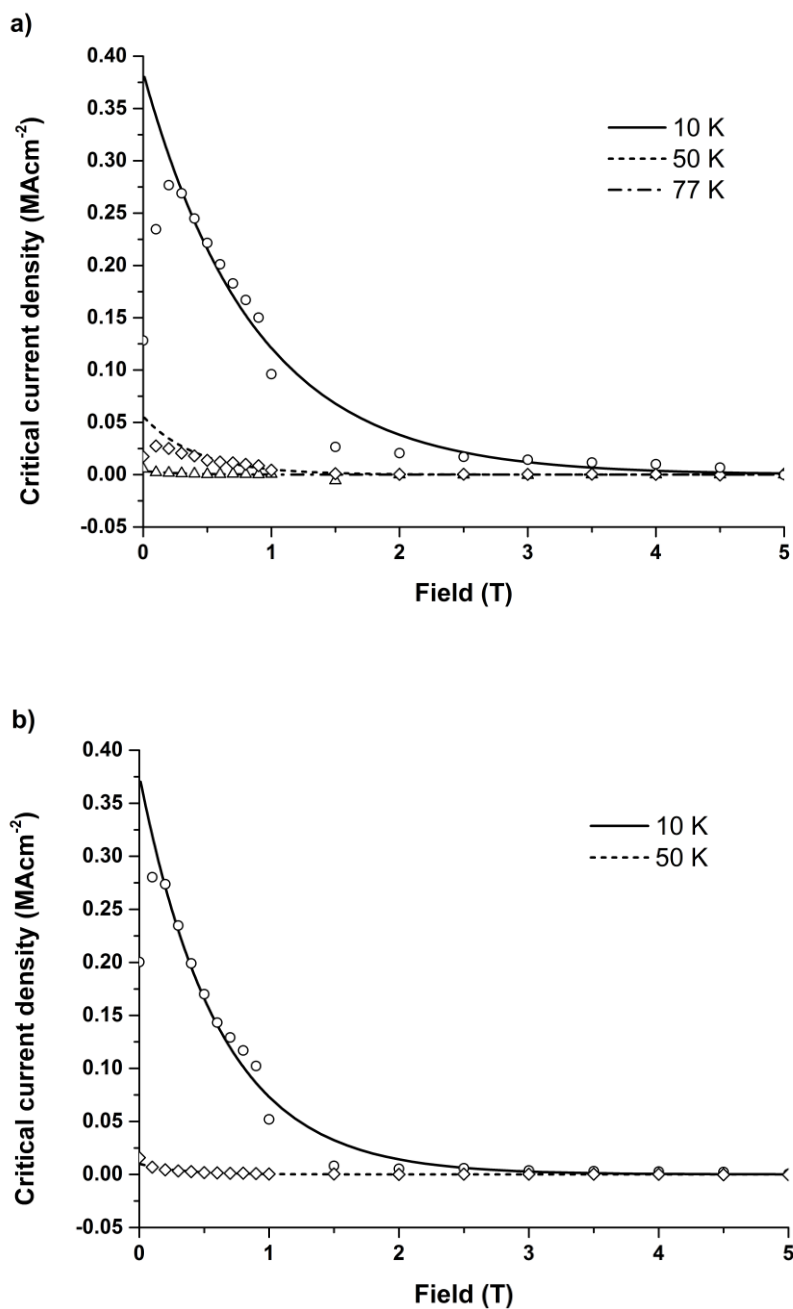


Figure S3. SQUID magnetometry showing critical current density of a) the Y123 foam, and b) the Y123 layered samples. Exponential best fit lines are shown for each dataset.

### Supporting Information References

- (1) N. I. Kovtyukhova, P. J. Ollivier, B. R. Martin, T. E. Mallouk, S. A. Chizhik, E. V Buzaneva, and A. D. Gorchinskiy, *Chem. Mater.*, 1999, **11**, 771.
- (2) W. L. Li, K. Lu, and J. Y. Walz, *Int. Mater. Rev.*, 2012, **57**, 37
- (3) P. Bean, *Phys. Rev. Lett.*, 1962, **8**, 250.