Electronic Supplementary Material (ESI) for Journal of Materials Chemistry A.

Supplementary Information for

Bulk monolithic electrodes enabled by surface mechanical attrition treatment-facilitated dealloying

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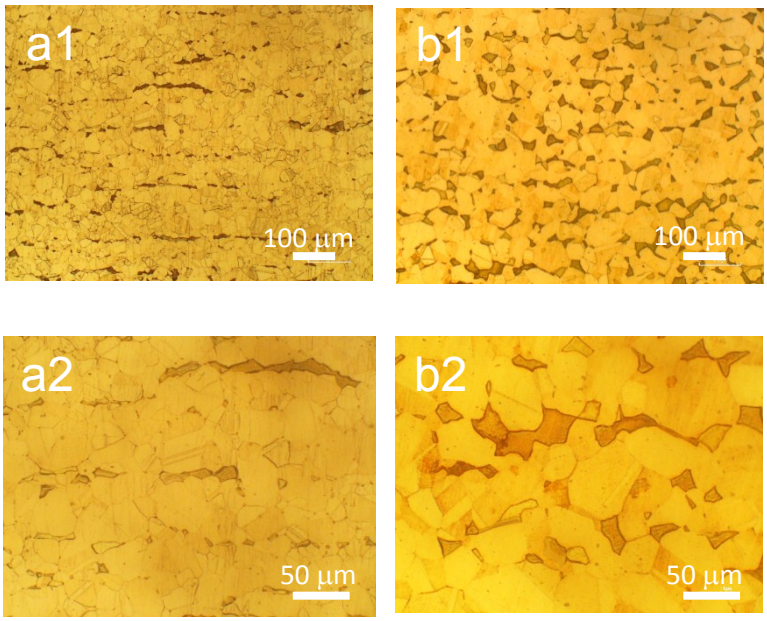

Fig. S1. Optical micrographs of the CuZn alloys as-received (a1, a2) and after annealed at 600 ˚C for 1 hr (b1, b2). It can be seen that the CuZn ally is a 2-phase system. The two phases in the as-received sample displayed elongated and oriented grains, possibly resulted from the cold work treatment during manufacturing, whereas the thermal annealing restored homogeneous and uniform grain distribution for both phases.

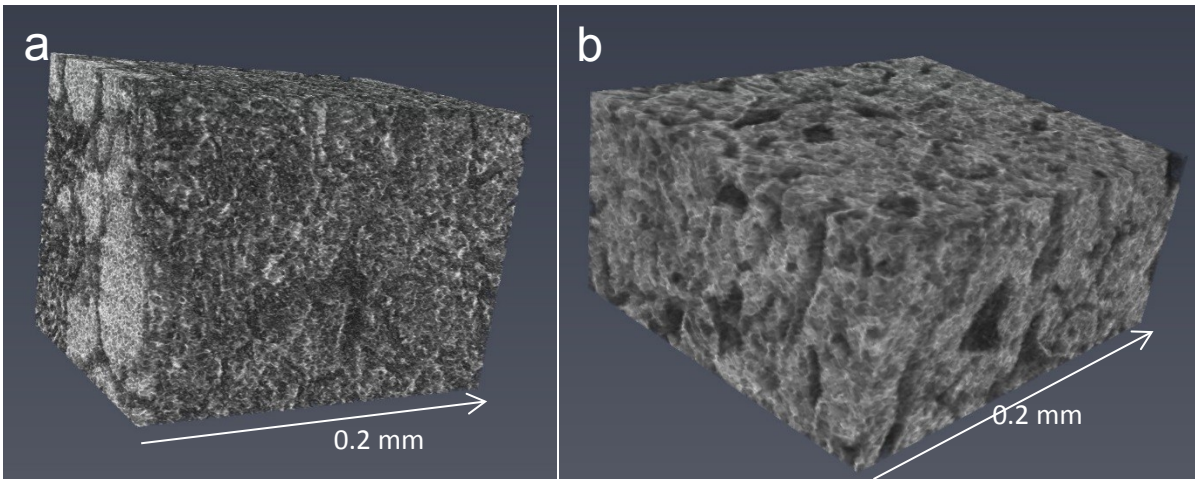

Fig. S2. X-ray synchrotron tomography images of bulk porous Cu structures that was SMATed for 0 (a) and 12 (b) min and dealloyed for 7 hr. Note that the darker regions in b is the dealloyed β phase.

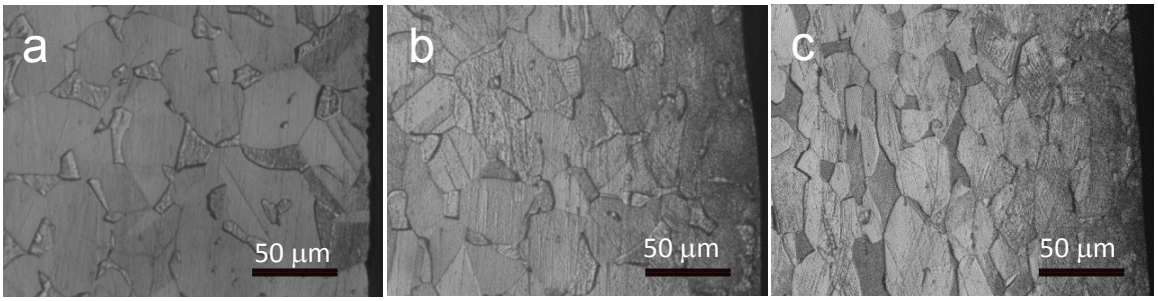

Fig. S3. Cross-sectional optical metallographic images of the as-annealed CuZn alloy after subjection to SMAT for 0 (a), 1 (b), and 12 (c) min. Reduced grain sizes, denser grain boundaries, and accumulated defects (e.g., slip or twinning boundaries) are clearly observed after the SMAT.

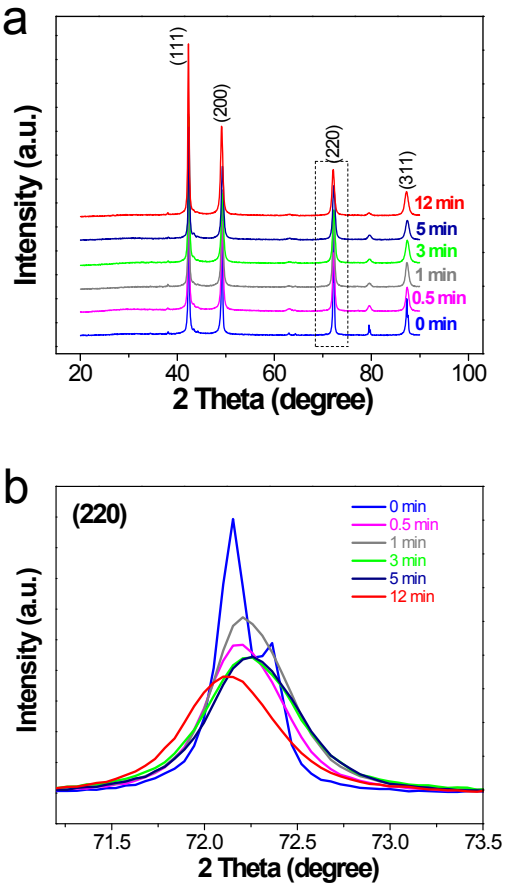

Fig. S4. XRD patterns collected on the CuZn samples that were SMATed for different time duration.

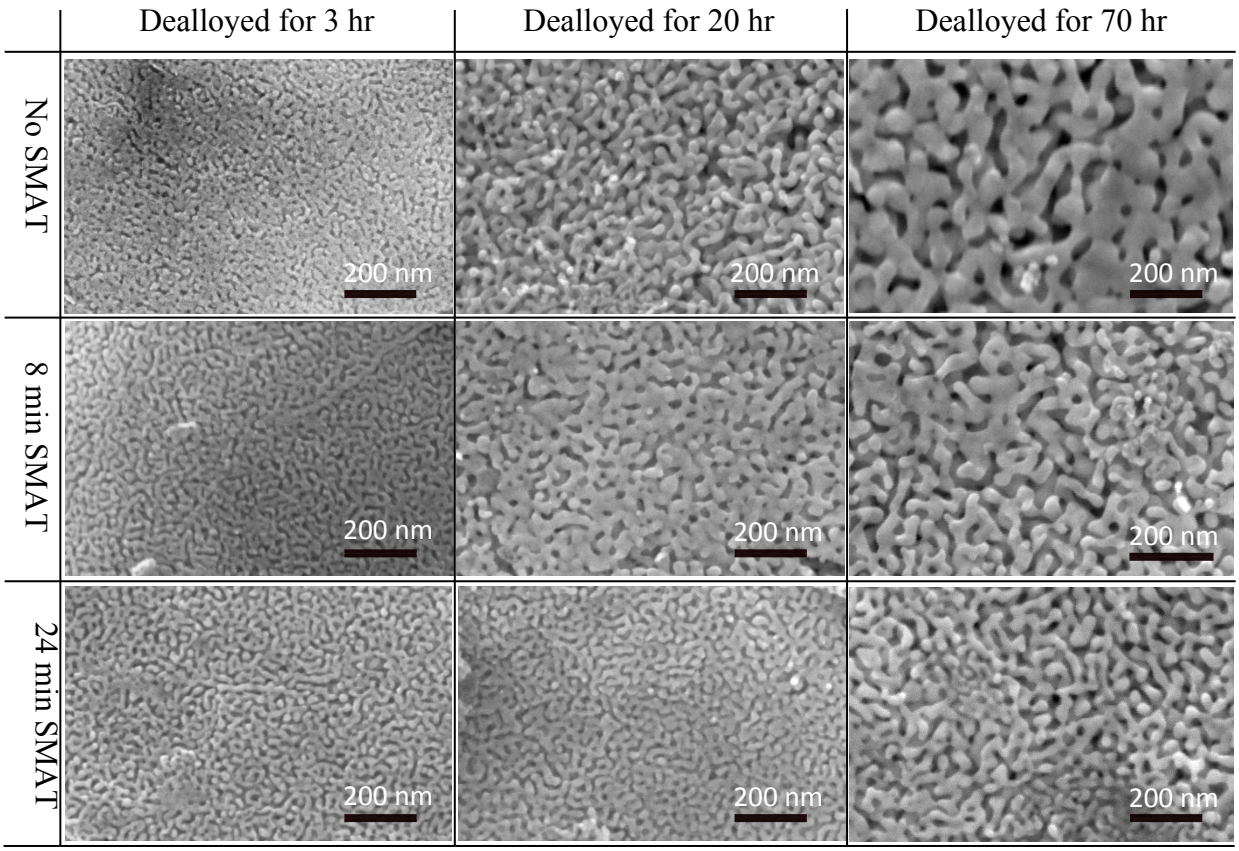

Fig. S5. SEM images of nanoporous gold fabricated from the commercial AuAg pellet (Kurt J. Lesker, $Au_{44}Ag_{56}$ by weight, 99.99% pure, 10 mm diameter × 10 mm long). The AuAg pellet was wire cut into 0.5 mm thick disks, polished, SMATed, and dealloyed in 67% HNO_3 at room temperature. The SMAT effects observed on the AuAg alloy system is consistent with the CuZn alloy system, e.g., anti-coarsening enabled.

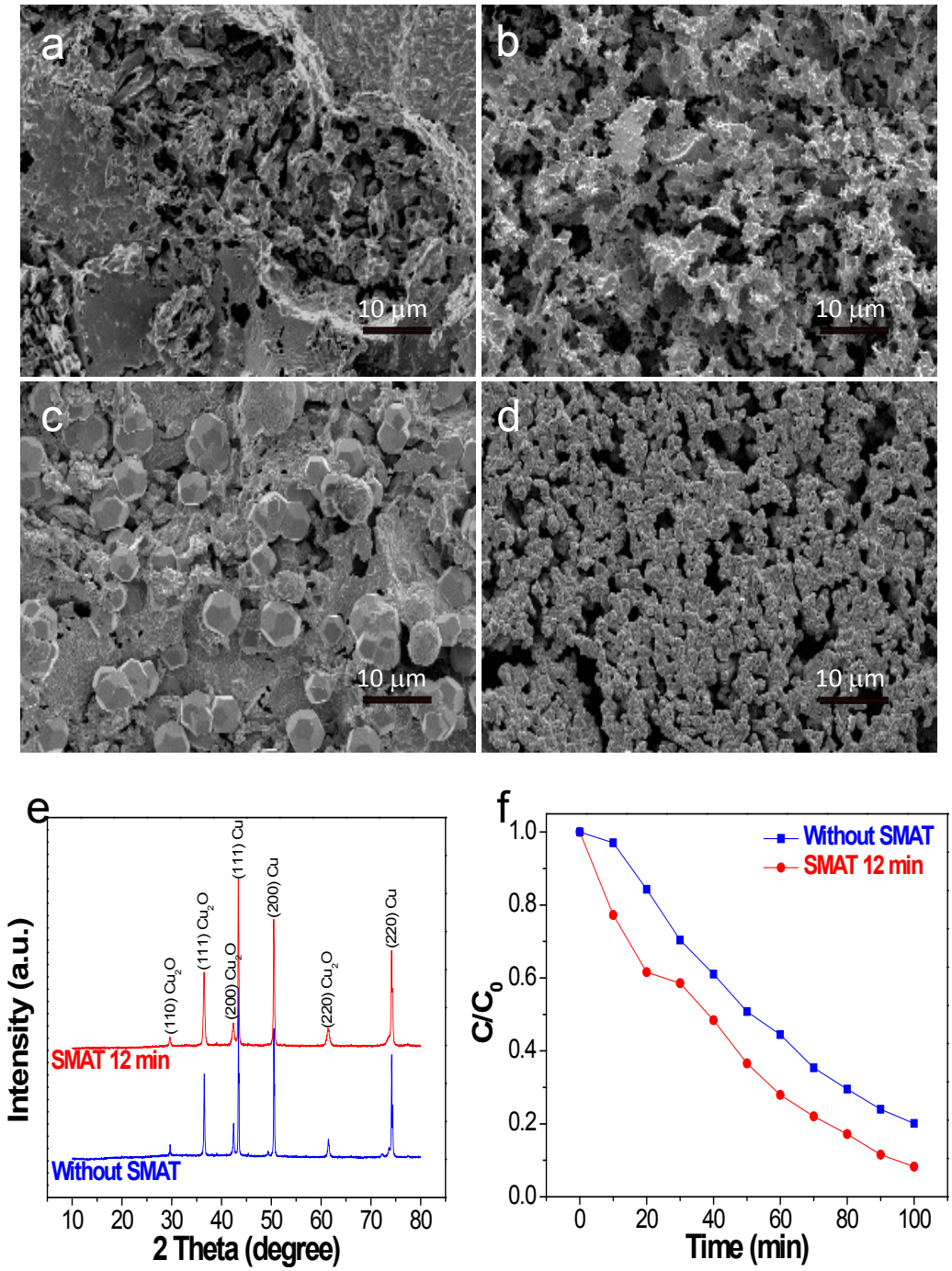

Fig. S6. a-d) SEM images of the porous Cu frameworks that was pre-SMATed for 0 (a,c) and 12 (b,d) min and dealloyed for 7 hours, before (a,b) and after (c,d) coated with Cu_2O. e) XRD

patterns of the Cu_2O-coated Cu frameworks whose SEM images were shown in c, d. f) Ratio of the actual and initial concentrations (c/c_0) of methyl blue as a function of the UV illumination time measured on the sample whose SEM images were shown in c, d. The Cu_2O coating was fabricated by soaking the dealloyed porous Cu frameworks (dealloyed for 7 hr) in an aqueous solution of copper (II) sulfate (Aldrich, 2.5 mM), ammonium nitrite (Aldrich, 1.25 mM), and ammonium hydroxide (Riedal–Dehaën, 2.7%) at 90 °C for 20 min.

Table S1. Performance comparison with some recent bulk framework-based supercapacitor electrodes

Material system	Area-specific capacitance (F cm^{-2})	Volume-specific capacitance (F cm^{-3})	Reference
NiOOH@Cu film (250 μm thick)	11.2 @ 6 mA cm^{-2} 10.5 @ 12 mA cm^{-2} 9.63 @ 20 mA cm^{-2}	448.8 @ 6 mA cm^{-2} 420.1 @ 12 mA cm^{-2} 385.2 @ 20 mA cm^{-2}	This study
CNT by CVD/MnO_2 by hydrothermal @ graphene foam by templating Ni foam (1 mm thick)	0.75 @ 5 mV s^{-1}		Energy Environ. Sci., 7, 3709 (2014)
$NiCo_2O_4$ nanoneedles @ Ni foam	3.12 @ 1.11 mA cm^{-2} 0.79 @ 11.12 mA cm^{-2}		Energy Environ. Sci. 5, 9453 (2012)
NiCuMnOOH oxyhydroxide by dealloying $Ni_{15}Cu_{15}Mn_{70}$ (at.%) alloy ribbons (20 μm thick)	1.2054 @ 0.5 mA cm^{-2}	627 @ 0.5 mA cm^{-2}	Angew. Chem. Int. Ed. 54, 8100 (2015)
Reduced graphene oxide-based hydrogel film (~ 350 μm thick)	0.071 @ 1 mA cm^{-2} 0.063 @10 mA cm^{-2} 0.0547 @ 100 mA cm^{-2}		Adv. Mater. 27, 4469 (2015)
$Ni(OH)_2$ by electrodeposition/CoO nanowalls by hydrothermal @ Ni foam (~ 0.1 mm thick)	11.5 @ 5 mA cm^{-2} 6.49 @ 40 mA cm^{-2}		Adv. Mater. 24, 4186 (2012)
$Ni(OH)_2$ by chemical bath deposition/CNT by CVD @ Ni foam	16 @ 2.5 mA cm^{-2}		Adv. Funct. Mater. 22, 1272 (2012)
Porous Ni film (50 μm) by dealloying NiMn film	1.40 @ 1 mA cm^{-2}	334 @ 1 mA cm^{-2}	J. Power Sources 247, 896 (2014)
Cobalt oxide nanobrush-graphene @ $Ni_xCo_{2x}(OH)_{6x}$ by hydrothermal, two-step calcination and two-step electrodeposition	5.1 @ 1 A g^{-1} 4.96 @ 2 A g^{-1} 4.7 @ 5 A g^{-1} 4.66 @ 10 A g^{-1} 4.23 @ 20 A g^{-1}		Nano Lett. 15, 2037 (2015)
CoO/Ppy nanowire array @ Ni foam (~1.1 mm thick)	4.43 @ 1 mA cm^{-2} 2.51 @ 5 mA cm^{-2} 2.13 @ 10 mA cm^{-2} 1.79 @ 20 mA cm^{-2} 1.28 @ 50 mA cm^{-2}		Nano Lett. 13, 2078 (2013)
MnO_2/CNT ink @ sponge (~ 1 mm thick) by dip coating and electrodeposition	0.0009 @ 1 mV s^{-1}		Nano Lett. 11, 5165 (2011)
RuO_2 nanoparticles @ graphene-CNT hybrid foam (0.5 mm thick) by CVD and sol-gel synthesis	1.11 @ 1 mA cm^{-2}		Sci. Rep. 4, 4452 (2014)