## Supporting Information

## Sol-gel Autocombustion Synthesis of Nanocrystalline

## **High-entropy Alloys**

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**Supplementary Figure S1.** XRD patterns of the CoCrCuNiAl HEA with a fuel–oxidant ratio of 1:1 and the CoCrCuNiAl HEA powder annealed at 1000 °C. The synthesized CoCrCuNiAl HEA is composed of FCC and BCC solid solutions, and the annealed HEA shows the same structure except for an increase in the diffraction peaks.  $Cr_3Ni_2$  phase is detected after annealing at 1,000 °C because of the decomposition of the HEAs at the high temperature. No oxides is detected after annealing at 1000 °C, indicating that there is no amorphous oxides in the sol–gel combustion product.

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Supplementary Figure S2. Additional information on the CoCrCuNiAl HEA prepared via mechanical alloying: a) TEM image, b) XRD pattern. The ball-milled alloy powder consists of simple solid solutions of the FCC and BCC structure with a particle size of about 3 µm.



Supplementary Figure S3. XRD patterns of the combustion product of the gels with fuel-oxidant ratios of 0.8:1 and 0.9:1 in the flowing hydrogen atmosphere.



Supplementary Figure S4. TEM images showing the combustion products prepared from sols with a fuel-oxidant ratio of a) 0.8:1, b) 0.9:1, c) 1.1:1 and d) 1:2:1, respectively.



Supplementary Figure S5. XRD results obtained for the CoCuNi and CoCrCuNI alloys with prepared from sols with different fuel-oxidant ratios. The CoCuNi and CoCrCuNI alloys prepared from the sols with the lowest fuel-oxidant ratio were selected according to the optimal fuel-oxidant ratio determined for the CoCrCuNiAl HEA.