Supporting Information

Sub-Ångstrom-scale structural variations in high-entropy oxides

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Table S1 Crystallographic data for the four types of HEOs from refinements of their powder XRD patterns.

Samples	(Mg,Co,Ni,Cu,Zn)O	$(Yb,Tb,Gd,Dy,Er)_2Ti_2O_7$	$(Cr,Mn,Fe,Co,Ni)_3O_4$	La(Cr,Mn,Fe,Co,Ni)O_3
Crystal system	Cubic			Orthorhombic
Space group	$Fm\bar{3}m$	$Fd\bar{3}m$	$Fd\bar{3}m$	$Pbnm$
Lattice Parameter [Å]	4.2365(2)	10.1117(2)	8.3477(3)	*a*=5.4747(2) *b*=5.5172(2) *c*=7.7542(4)
Volume [Å^3]	76.034(7)	1033.88(3)	581.70(4)	234.21(2)
Calculated Density [g cm^{-3}]	6.13	6.88	5.30	6.89
R_p	3.53%	7.42%	5.57%	8.37%
Bragg R-factor	2.83%	6.20%	7.50%	7.18%
Goodness of Fit [χ^2]	5.58	5.64	4.75	4.85

Table S2 Refined atomic coordinates for the four types of HEOs from their powder XRD patterns.

(Mg,Co,Ni,Cu,Zn)O				
atom	Wyckoff site	*x*	*y*	*z*
M	4*a*	0	0	0
O	4*b*	0.5	0.5	0.5
M = Mg, Co, Ni, Cu, and Zn with occupancy = 0.2 for each.				
$(Yb,Tb,Gd,Dy,Er)_2Ti_2O_7$				
atom	Wyckoff site	*x*	*y*	*z*
M	16*c*	0	0	0
Ti	16*d*	0.5	0.5	0.5
O1	8*a*	0.125	0.125	0.125
O2	48*f*	0.4277(7)	0.125	0.125
M = Yb, Tb, Gd, Dy, and Er with occupancy = 0.2 for each.				
$(Cr,Mn,Fe,Co,Ni)_3O_4$				
atom	Wyckoff site	*x*	*y*	*z*
*M*1	8*a*	-0.125	-0.125	-0.125
*M*2	16*d*	0.500	0.500	0.500
O	32*e*	0.2597(4)	0.2597(4)	0.2597(4)
*M*1 =Cr, Mn, Fe, Co, and Ni with occupancy = 0.2 for each; *M*2 =Cr, Mn, Fe, Co, and Ni with occupancy = 0.2 for each.				
$La(Cr,Mn,Fe,Co,Ni)O_3$				
atom	Wyckoff site	*x*	*y*	*z*
M	4*b*	0.5	0	0
La	4*c*	0.001(2)	0.0141(5)	0.25
O1	4*c*	-0.04(1)	0.466(6)	0.25
O2	8*d*	0.289(3)	0.282(3)	0.037(2)
M = Cr, Mn, Fe, Co, and Ni with occupancy = 0.2 for each.				

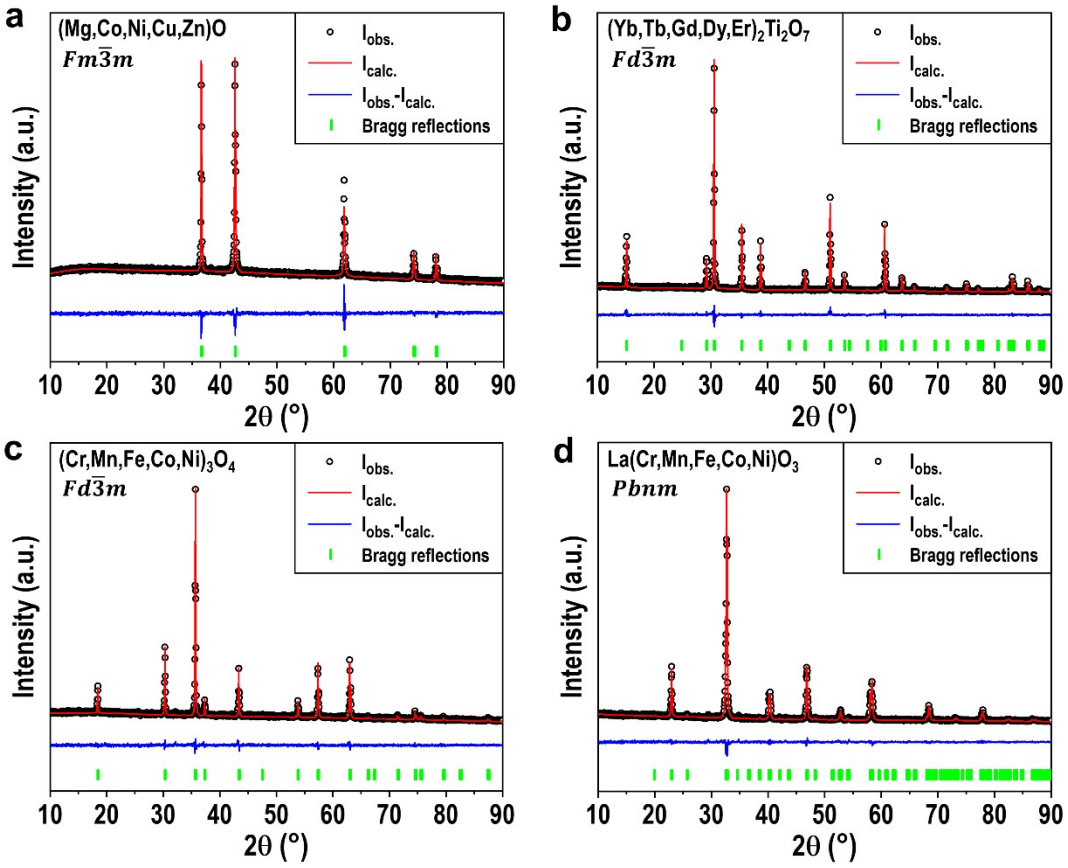

Fig. S1 XRD patterns for sintered samples of (**a**) rock-salt type (Mg,Co,Ni,Cu,Zn)O, (**b**) pyrochlore type $(Yb,Tb,Gd,Dy,Er)_2Ti_2O_7$, (**c**) spinel type $(Cr,Mn,Fe,Co,Ni)_3O_4$, and (**d**) perovskite type $La(Cr,Mn,Fe,Co,Ni)O_3$.

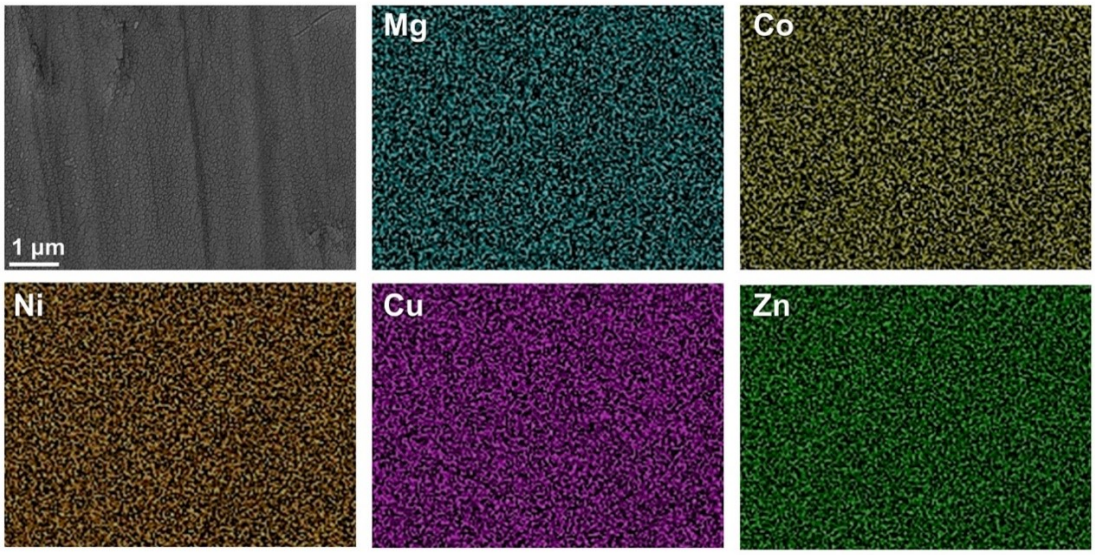

Fig. S2 EDS-SEM elemental maps of Mg, Co, Ni, Cu and Zn for rock-salt type (Mg,Co,Ni,Cu,Zn)O HEO.

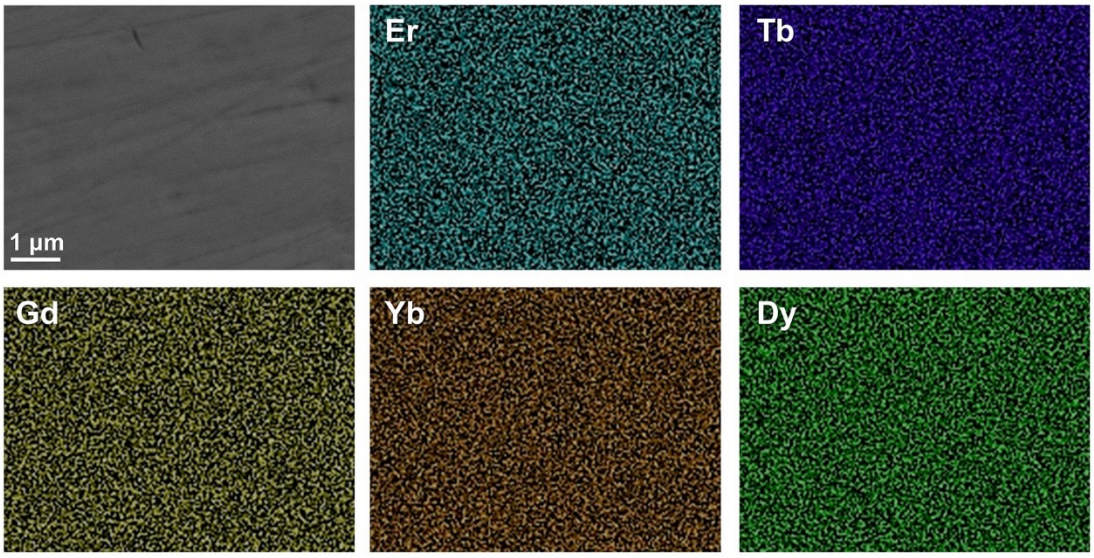

Fig. S3 EDS-SEM elemental maps of Er, Tb, Gd, Yb and Dy for pyrochlore type $(Yb,Tb,Gd,Dy,Er)_2Ti_2O_7$ HEO.

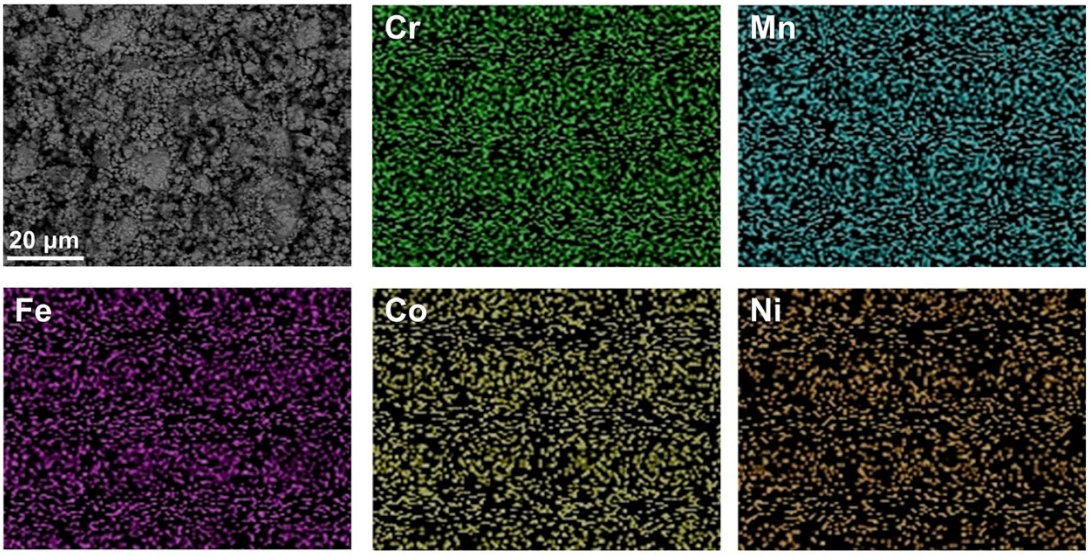

Fig. S4 EDS-SEM elemental maps of Cr, Mn, Fe, Co and Ni for spinel type $(Cr,Mn,Fe,Co,Ni)_3O_4$ HEO.

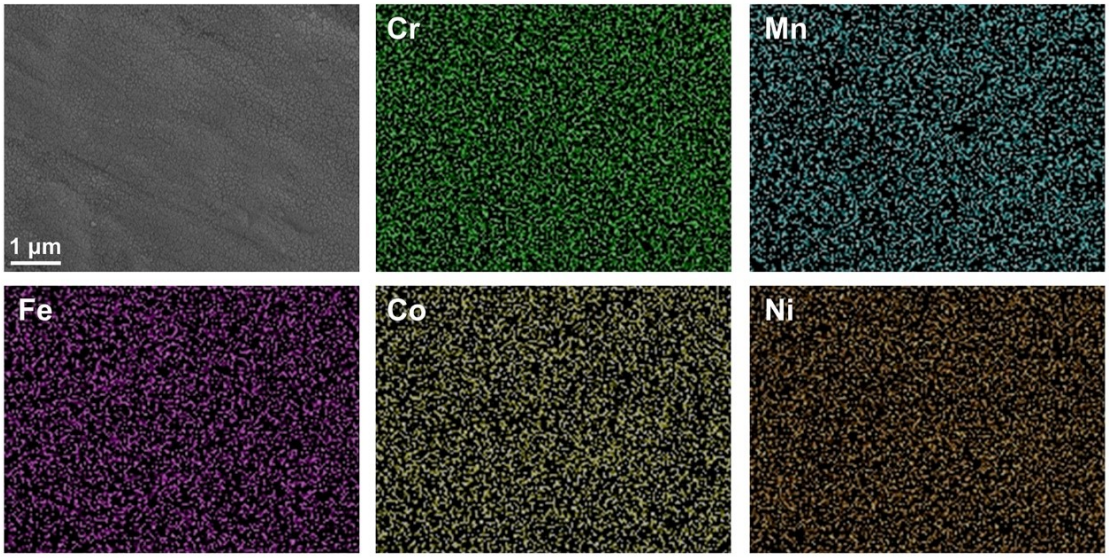

Fig. S5 EDS-SEM elemental maps of Cr, Mn, Fe, Co and Ni for perovskite type $La(Cr,Mn,Fe,Co,Ni)O_3$ HEO.

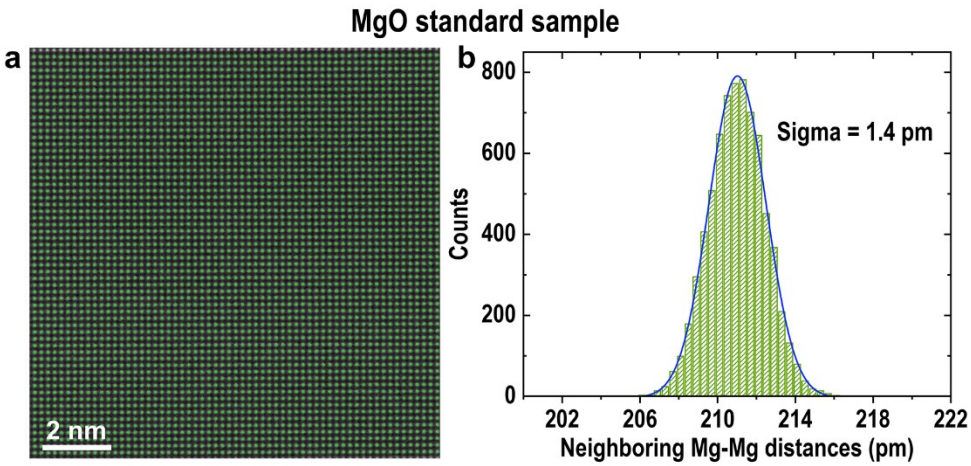

Fig. S6 Precision of atomic position fits estimated from the MgO standard sample. (**a**) The positions of atomic columns obtained by 2D Gaussian fits. (**b**) Histogram of distances between neighboring Mg atomic columns. The Gaussian fits for this histogram show Sigma (precision) is 1.4 pm.

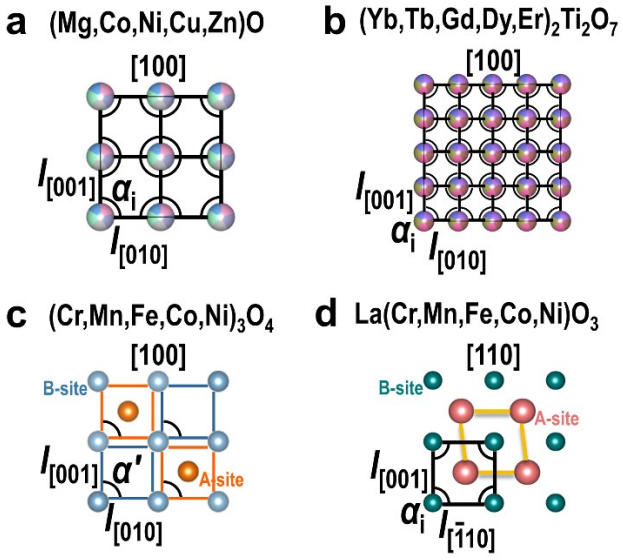

Fig. S7 Quantification methods of aspect ratios and inner angle variations from 90° for all quadrangle units in HAADF images of the four types of HEOs. Side lengths and inner angles for a quadrangle unit in (**a**) rock-salt type (Mg,Co,Ni,Cu,Zn)O and (**b**) pyrochlore type (Yb,Tb,Gd,Dy,Er)$_2$Ti$_2$O$_7$ HEOs. The aspect ratio for a quadrangle unit is calculated based on $\frac{l_{[001]}^{left} + l_{[001]}^{right}}{l_{[010]}^{up} + l_{[010]}^{down}}$. (**c**) Side lengths and inner angles for a quadrangle unit in the spinel type (Cr,Mn,Fe,Co,Ni)$_3$O$_4$ HEO. The aspect ratio and inner angle deviation parameter from 90° for a quadrangle unit are calculated based on $\frac{l_{[001]}}{l_{[010]}}$ and $\alpha = \alpha'$**-90**, respectively. (**d**) Side lengths and inner angles for a quadrangle unit in the perovskite type La(Cr,Mn,Fe,Co,Ni)O$_3$. The aspect ratio and inner angle deviation parameter from 90° for a quadrangle unit are calculated based on $\frac{l_{[001]}^{left} + l_{[001]}^{right}}{l_{[\bar{1}10]}^{up} + l_{[\bar{1}10]}^{down}}$ and $\frac{\sum_{i}^{4} |\alpha_i \text{ - } \mathbf{90}|}{4}$, respectively.

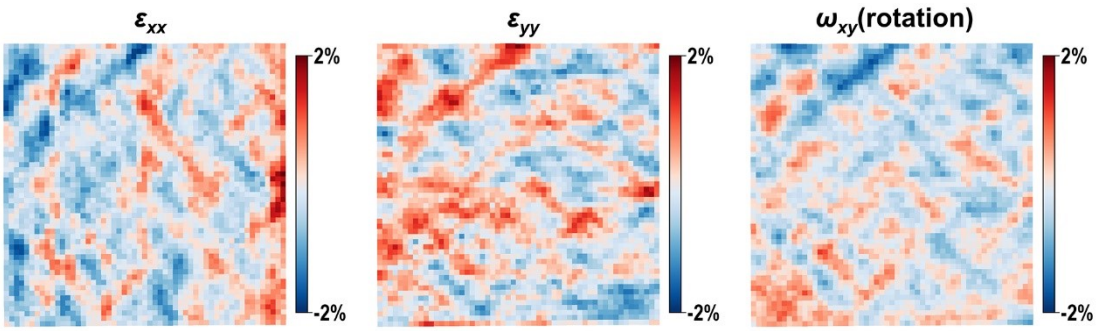

Fig. S8 Real-space strain analysis for Fig. 2(a) with uniaxial strain component ε_{xx} and ε_{yy} and rotation ω_{xy} maps for the rock-salt type (Mg,Co,Ni,Cu,Zn)O HEO.

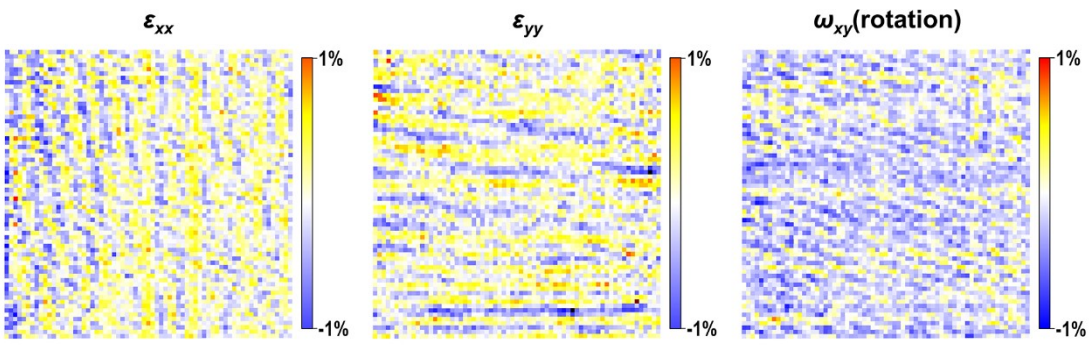

Fig. S9 Real-space strain analysis for Fig 3(a) with uniaxial strain component ε_{xx} and ε_{yy} and rotation ω_{xy} maps for pyrochlore type $(Yb,Tb,Gd,Dy,Er)_2Ti_2O_7$ HEO.