Supporting information

Heusler alloys: a group of novel catalysts

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Table S1. List of prepared Heusler alloys and related information. S and α are ordering factors based on Webster's model,[1] and SD values are their standard deviations. Details about S, α, and SD are described in the main text. The presence of an extra phase was judged from the XRD patterns; "yes" means that extra peaks can be clearly seen but their intensities are negligible, and "almost no" means that presence of extra phases can be confirmed but their peaks are unclear. The product of the surface area and the amount of catalyst corresponds to 0.027 m^2. As-arc-melted alloys were annealed for homogenization followed by chemical ordering under the listed conditions.

Material	S	SD	α	SD	Extra phase	Surface area [10^{-2} m^2 g^{-1}]	Amount of catalyst [mg]	Annealing to homogenize	Annealing to order
Fe_2TiSn	1.0	–	0.05	–	yes	7.1	378	850 °C 72 h	600 °C 144 h
Co_2TiAl	0.97	0.04	0.14	0.02	no	11	237	1000 °C 72 h	500 °C 24 h
Co_2TiGe	1	–	0.01	–	no	8.4	320	1000 °C 72 h	600 °C 144 h
Co_2TiSn	0.99	0.01	0.01	<0.01	yes	7.0	387	850 °C 72 h	500 °C 48 h
Co_2MnSi	1.00	0.04	0.06	<0.01	no	8.8	307	1000 °C 40 h	600 °C 100 h
Co_2MnGe	1.0	–	0.05	–	no	7.2	373	1000 °C 72 h	600 °C 144 h
Co_2MnSn	0.97	0.01	0.05	<0.01	no	5.6	478	800 °C 120 h	600 °C 144 h
Co_2FeGe	0.97	0.02	0.04	<0.01	yes	6.2	436	800 °C 72 h	500 °C 48 h
Ni_2TiAl	0.98	0.01	0.01	<0.01	no	13	203	1000 °C 72 h	600 °C 144 h
Ni_2TiSn	1.0	–	0.00	–	yes	9.5	284	850 °C 120 h	600 °C 144 h
Ni_2MnSn	1.0	–	0.0	–	almost no	8.9	304	800 °C 120 h	500 °C 48 h
Cu_2TiAl	0.95	<0.01	0.00	<0.01	almost no	12	223	850 °C 72 h	500 °C 48 h
Commercial Co powder						6.4	422		

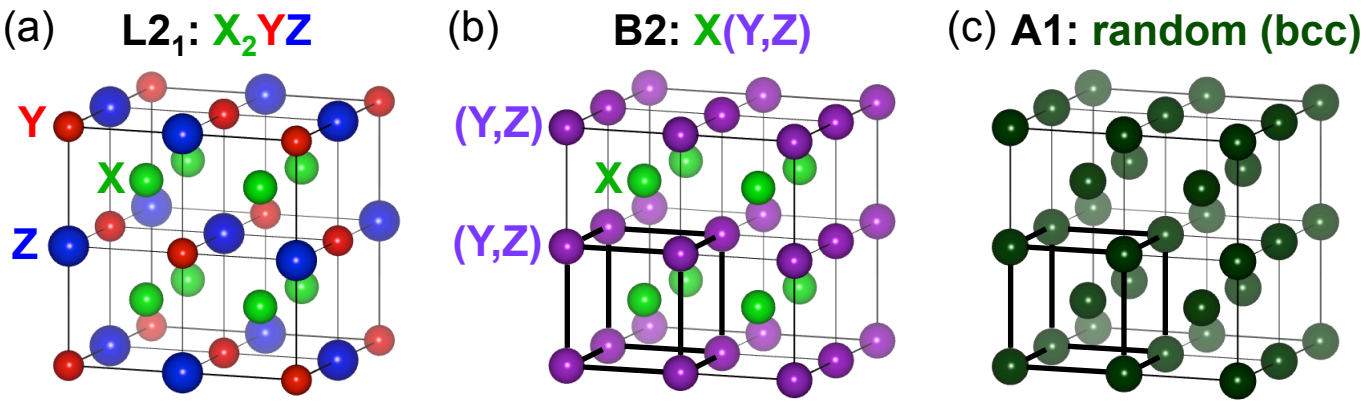

Figure S1. Crystal structures of (a) $L2_1$ phase and typical disordered phases in Heusler alloys: (b) B2 and (c) A1. B2 phase is disordered between Y and Z but ordered between X and (Y, Z). A1 phase is a completely disordered (random) structure (bcc structure). Unit cells of B2 and A1 are shown by thicker lines in (b) and (c). Figures were drawn using VESTA.[2]

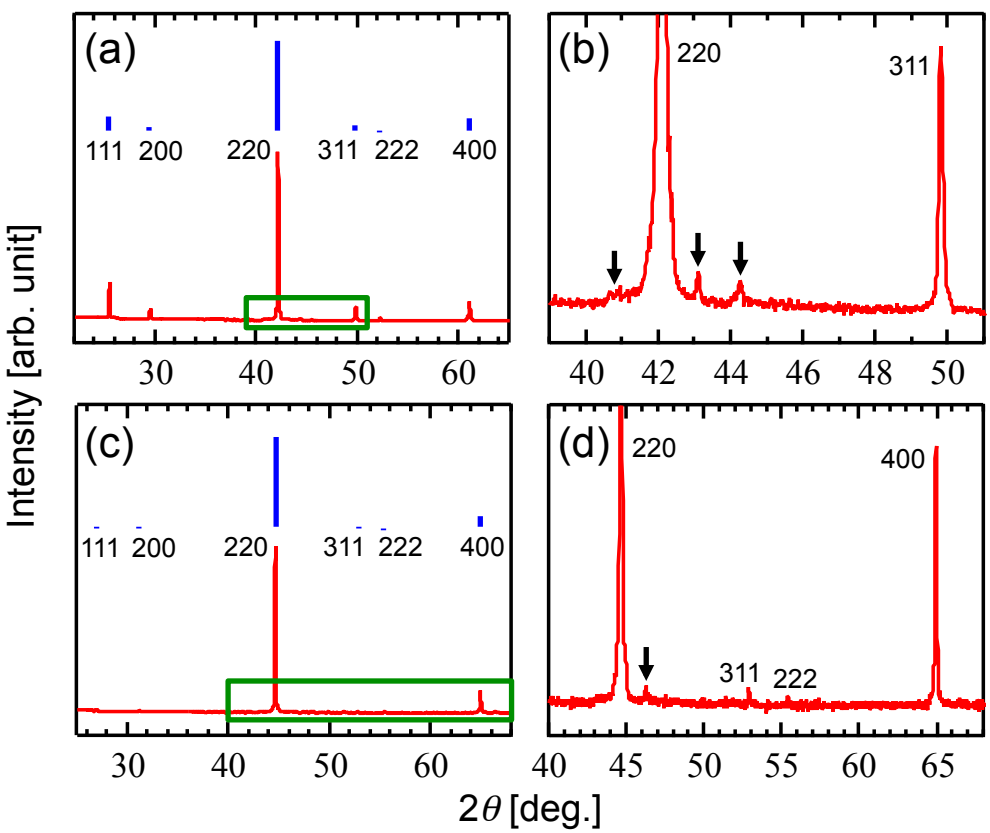

Figure S2. XRD patterns of (a, b) Co_2TiSn and (c, d) Co_2FeGe. (b) and (d) are magnified images in areas enclosed by green boxes in (a) and (c), respectively. Reflection indices are based on $L2_1$ structure. Blue bars in (a) and (c) are calculated peaks for each material. Arrows in (b) and (d) indicate peaks of extra phases.

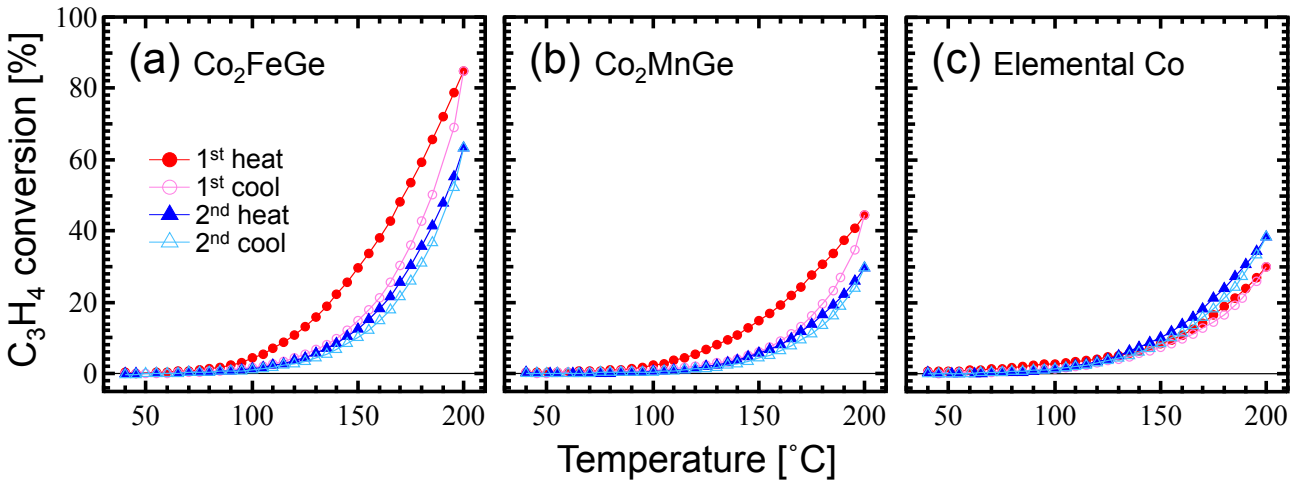

Figure S3. Conversion of C_3H_4 in hydrogenation of propyne (C_3H_4) by (a) Co_2FeGe, (b) Co_2MnGe, and (c) elemental Co. Data points were divided among thermal processes as shown by legend in (a). Heating: filled symbols, Cooling: open symbols, 1st cycle: red and pink circles, 2nd cycle: deep and pale blue triangles.

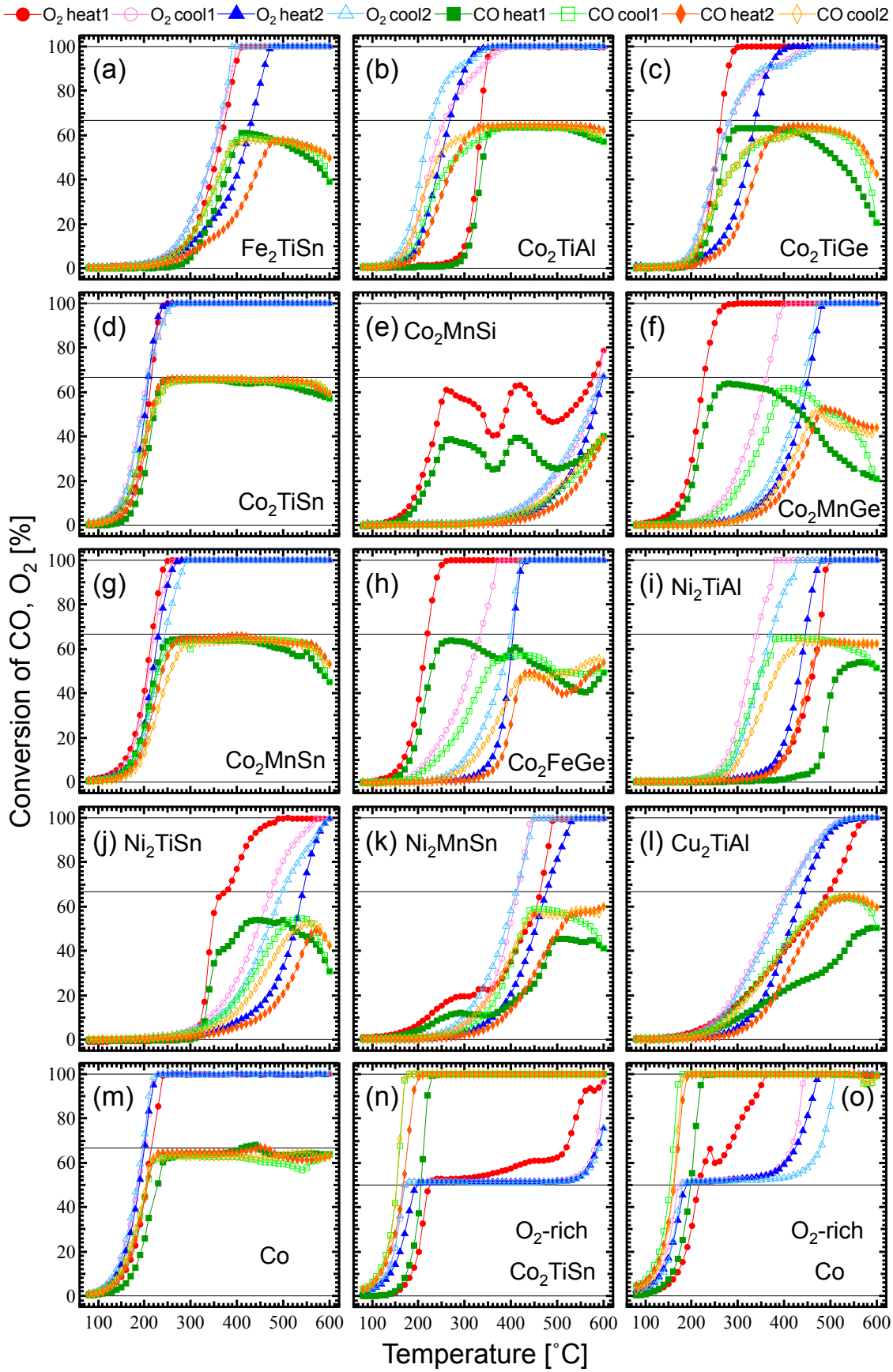

Figure S4. Conversion of CO and O_2 in oxidation of carbon monoxide. Material names are shown in figure. Reactants were CO-rich (CO : O_2 = 3 : 1) for (a)–(m) and O_2-rich (CO : O_2) = (1 : 1) for (n,o). Data points were divided among thermal processes as shown by legend at top of figure. Heating: filled symbols, Cooling: open symbols, O_2 conversion: red and pink circles (1st cycle) and deep and pale blue triangles (2nd cycle), CO conversion: deep and pale green squares (1st cycle) and deep and pale orange diamonds (2nd cycle). Horizontal lines at conversions of 0%, 100%, and 66.7% (ideal value for CO) in (a)–(m) and 50% (ideal value for O_2) in (n,o) are guides for the eye.

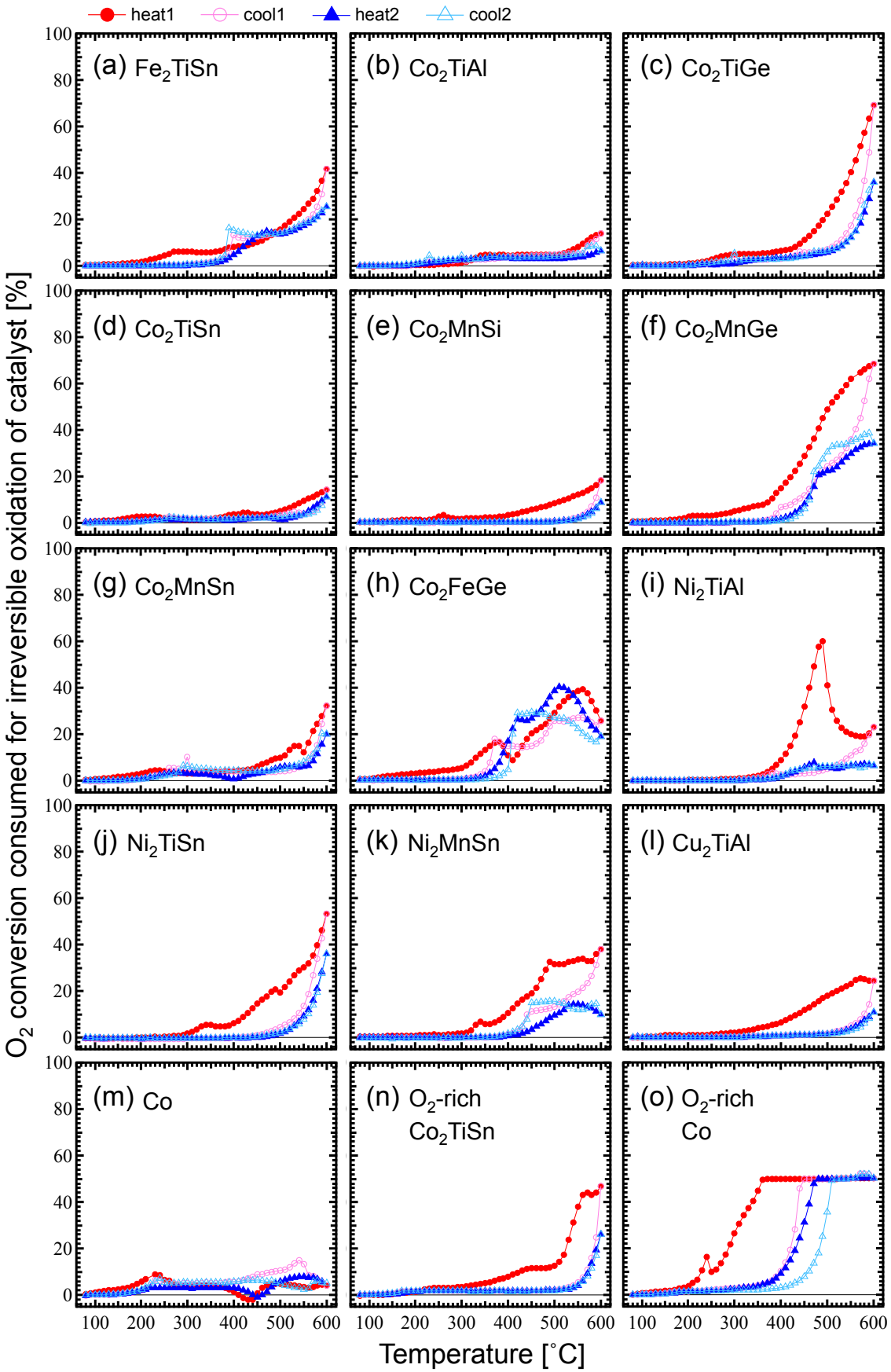

Figure S5. Conversion of O_2 consumed for irreversible oxidation of catalyst in oxidation of carbon monoxide, calculated from Fig. S4. Reactants were CO-rich (CO : O_2 = 3 : 1) for (a)–(m) and O_2-rich (CO : O_2) = (1 : 1) for (n,o). Figure identifiers correspond to those in Fig. S4. Symbols also correspond to those in Fig. S4, as shown by legend at top of figure. Heating: filled symbols, Cooling: open symbols, 1st cycle: red and pink circles, 2nd cycle: deep and pale blue triangles.

References

1. Webster, P.J.; Ziebeck, K.R.A. Magnetic and chemical order in Heusler alloys containing cobalt and titanium. *J. Phys. Chem. Solids* **1973**, *34*, 1647–1654.

2. Momma, K.; Izumi, F. *VESTA 3* for three-dimensional visualization of crystal, volumetric and morphology data. *J. Appl. Crystallogr.* **2011**, *44*, 1272–1276.