Supporting Information

Part 1: Structures of Special Quasirandom Structure and Ordered Bulk Periodic Cells

Table S1. Lattice constants and angles of unrelaxed alloy unit cells. The lattice constants are given in units of the distance between a transition metal and an oxygen atom (denoted TM-O). A Special Quasirandom Structure (SQS) cell that mimics random alloy correlation functions up through nearest-neighbor (next-nearest-neighbor) interactions is denoted with * (**).

Unit cell type	Alloying percentage	Lattice constants [TM-O distance]	Lattice angles (deg)
SQS cell*	0.25, 0.5, 0.75	$(\sqrt{6}, \sqrt{2}, \sqrt{22})$	(90,100,90)
SQS cell**	0.5	$(\sqrt{2}, 2\sqrt{2}, 4)$	(90,90,90)
Ordered cells	0.5	$(\sqrt{6}, \sqrt{6}, \sqrt{6})$	(33.6,33.6,33.6)

Table S2. Fractional coordinates of atoms in SQS bulk cells that mimic random alloy correlation functions up through nearest-neighbor interactions (before relaxation). Atom type *M = Mg, Mn, Ni,* or *Zn.* The total spin of each atom is indicated (atoms *O, Mg,* and *Zn* have none). The coordinates of the oxygen atoms are the same in all alloying concentrations and therefore omitted at alloying fraction *x=0.5*. A cell at alloying fraction *x=0.75* is the same as the one at *x=0.25* with *Fe* replacing *M* and vice-versa.

Alloying percentage	Atom type	Fractional coordinates
0.25	*O*	(1.125, 0, 0.375); (0.375, 0.5, 0.625); (0.875, 0, 0.625); (0.375, 0, 0.125); (0.625, 0.5, 0.375); (1.125, 0.5, 0.875); (0.625, 0, 0.875); (0.875, 0.5, 0.125);
	Fe (positive spin)	(0.5, 1, 0.5); (1, 1, 1); (0.5, 0.5, 1)
	Fe (negative spin)	(0.25, 1, 0.75); (0.25, 0.5, 0.25); (0.75, 1, 0.25)
	M (positive spin)	(1, 0.5, 0.5)
	M (negative spin)	(0.75, 0.5, 0.75)
0.5	*Fe* (positive spin)	(1, 0.5, 0.5); (0.25, 1, 0.75)
	Fe (negative spin)	(0.75, 0.5, 0.75); (0.25, 0.5, 0.25)
	M (positive spin)	(0.5, 0.5, 1); (0.75, 1, 0.25)
	M (negative spin)	(0.5, 1, 0.5); (1, 1, 1)

Table S3. Fractional coordinates of atoms in SQS bulk cells that mimic random alloy correlation functions up through next-nearest-neighbor interactions.

Alloying percentage	Atom type	Fractional coordinates
0.5	*O*	(1, 0.5, 0.25); (1.5, 0.25, 1); (1, 0.5, 0.75); (1.5, 0.25, 0.5); (1, 0, 0.25); (1.5, 0.75, 1); (1, 0, 0.75); (1.5, 0.75, 0.5);
	Fe (positive spin)	(0.5, 0.25, 0.75); (1, 1, 0.5)
	Fe (negative spin)	(1, 0.5, 0.5); (1, 1, 1)
	M (positive spin)	(0.5, 0.75, 0.25); (1, 0.5, 1)
	M (negative spin)	(0.5, 0.75, 0.75); (0.5, 0.25, 0.25)

Table S4. Fractional coordinates of atoms in several ordered bulk cells shown in Figure S1. The coordinates of the oxygen atoms are the same in all cells and therefore omitted from cell # 2, 3, and 4.

Alloying percentage	Atom type	Fractional coordinates
0.5 (cell # 1)	*O*	(0.125, 0.125, 0.125); (0.375, 0.375, 0.375); (0.625, 0.125, 0.125); (0.875, 0.375, 0.375); (0.125, 0.625, 0.125); (0.375, 0.875, 0.375); (0.625, 0.625, 0.125); (0.875, 0.875, 0.375); (0.125, 0.125, 0.625); (0.375, 0.375, 0.875); (0.625, 0.125, 0.625); (0.875, 0.375, 0.875); (0.125, 0.625, 0.625); (0.375, 0.875, 0.875); (0.625, 0.625, 0.625); (0.875, 0.875, 0.875)
	Fe (positive spin)	(0.5, 0, 0); (0, 0.5, 0); (0, 0, 0.5); (0.5, 0, 0.5)
	Fe (negative spin)	(0.75, 0.25, 0.25); (0.75, 0.75, 0.25); (0.25, 0.25, 0.75); (0.25, 0.75, 0.75)
	M (positive spin)	(0, 0, 0); (0.5, 0.5, 0); (0, 0.5, 0.5); (0.5, 0.5, 0.5)
	M (negative spin)	(0.25, 0.25, 0.25); (0.25, 0.75, 0.25); (0.75, 0.25, 0.75); (0.75, 0.75, 0.75)
0.5 (cell # 2)	*Fe* (positive spin)	(0.5, 0, 0); (0, 0.5, 0); (0, 0, 0.5); (0.5, 0, 0.5)
	Fe (negative spin)	(0.75, 0.25, 0.25); (0.25, 0.75, 0.75); (0.25, 0.25, 0.25); (0.75, 0.75, 0.75)
	M (positive spin)	(0, 0, 0); (0.5, 0.5, 0); (0, 0.5, 0.5); (0.5, 0.5, 0.5)
	M (negative spin)	(0.75, 0.75, 0.25); (0.25, 0.25, 0.75); (0.25, 0.75, 0.25); (0.75, 0.25, 0.75)
0.5 (cell # 3)	*Fe* (positive spin)	(0, 0.5, 0); (0, 0, 0.5); (0.5, 0, 0.5); (0, 0, 0)
	Fe (negative spin)	(0.75, 0.25, 0.25); (0.25, 0.75, 0.75); (0.25, 0.25, 0.25); (0.75, 0.75, 0.75)
	M (positive spin)	(0.5, 0, 0); (0.5, 0.5, 0); (0, 0.5, 0.5); (0.5, 0.5, 0.5)
	M (negative spin)	(0.75, 0.75, 0.25); (0.25, 0.25, 0.75); (0.25, 0.75, 0.25); (0.75, 0.25, 0.75)
0.5 (cell # 4)	*Fe* (positive spin)	(0, 0.5, 0); (0, 0, 0.5); (0.5, 0, 0.5); (0, 0, 0)
	Fe (negative spin)	(0.75, 0.25, 0.25); (0.75, 0.75, 0.25); (0.25, 0.25, 0.75); (0.25, 0.75, 0.75)
	M (positive spin)	(0.5, 0, 0); (0.5, 0.5, 0); (0, 0.5, 0.5); (0.5, 0.5, 0.5)
	M (negative spin)	(0.25, 0.25, 0.25); (0.25, 0.75, 0.25); (0.75, 0.25, 0.75); (0.75, 0.75, 0.75)

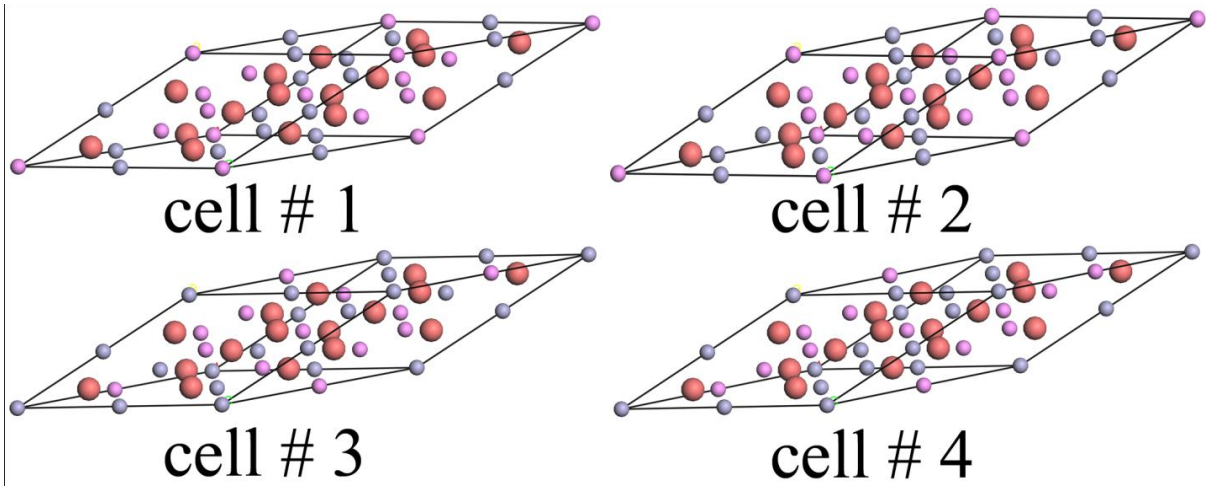

Figure S1. Ordered bulk unit cells. Red spheres represent *O*, blue spheres represent *Fe* and pink spheres represent *M*.

Part 2: Structures of Periodic Slab Unit Cells

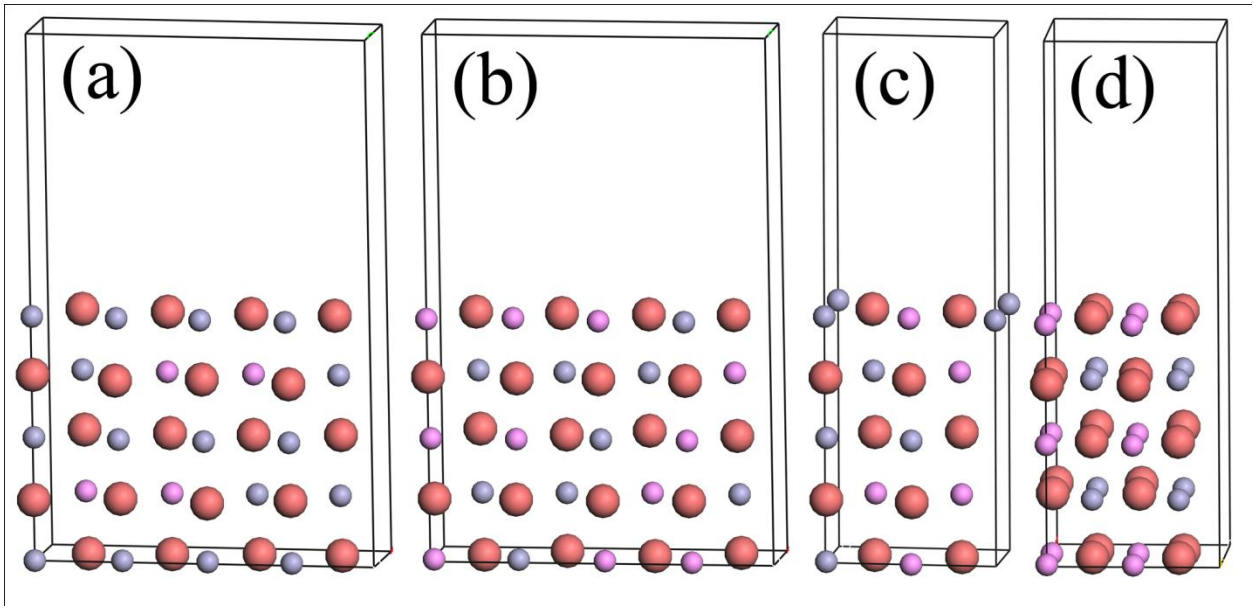

Figure S2. Periodic slab unit cells cleaved from a bulk SQS cell that mimics random alloy correlation functions up through: (a) nearest-neighbor interactions at alloying fraction *x=0.25*, (b) nearest-neighbor interactions at alloying fraction *x=0.5*, and (c) next-nearest-neighbor interactions at alloying fraction *x=0.5*. (d) displays an ordered periodic slab unit cell for *x=0.5*. A slab at alloying fraction *x=0.75* is the same as the one in (a) with *Fe* replacing *M*. The same color scheme is used as in Figure S1.

Part 3: Energetics of forming alloys

The alloy formation energy was calculated according to the following equation,

$$\Delta E = a \cdot E_t(Fe_{1-x}M_xO) - (1-x) \cdot E_t(FeO) - b \cdot x \cdot E_t(MO)$$

where E_t is a total energy and $M = Mg, Mn, Ni$ or Zn. The factor a=¼ that appears in front of the $Fe_{1-x}M_xO$ total energy is the number of atoms in the FeO primitive cell divided by the number of atoms in the alloy unit cells. The factor b that appears in front of the MO total energy is the number of atoms in the FeO primitive cell divided by the number of atoms in the MO unit cell. Antiferromagnetic rocksalt FeO has a 4-atom primitive cell, as does antiferromagnetic rocksalt MnO and NiO. The lowest energy phase of ZnO is wurtzite, which also has a 4-atom primitive cell. Hence, b=1 for MnO, NiO, and ZnO whereas b=2 for nonmagnetic rocksalt MgO since it has a 2-atom primitive cell. As seen in Figure S3, many of the energies of forming alloys are negative, suggesting that alloying may be thermodynamically favorable (we cannot make a definite conclusion without accounting for zero point energy and finite temperature entropic corrections, which is beyond the scope of this work). The total energy of wurtzite ZnO is lower by 0.1 eV/atom compared to the rocksalt phase. If the rocksalt phase had been taken instead, all energies in Figure S3 would have been negative.

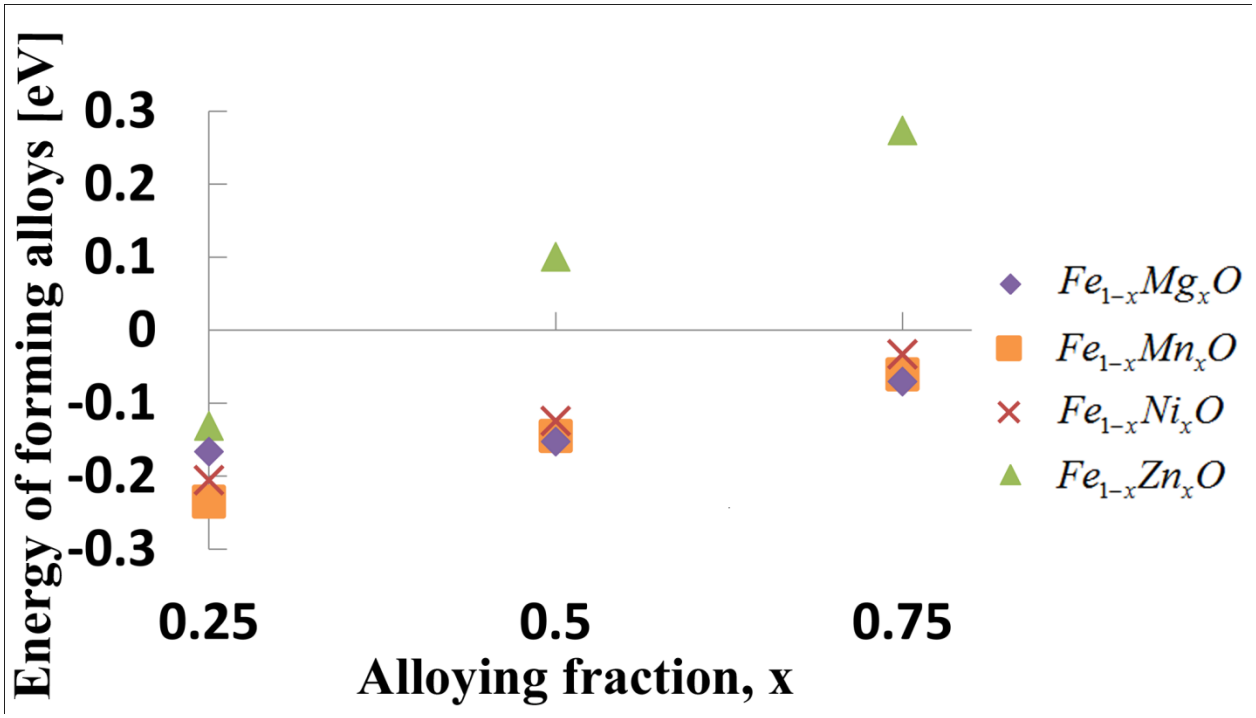

Figure S3. Energies of forming the alloys $Fe_{1-x}Mg_xO$, $Fe_{1-x}Mn_xO$, $Fe_{1-x}Ni_xO$, and $Fe_{1-x}Zn_xO$.