



Titre: Synthesis and characterization of (Al,Si)₃(Zr,Ti)-D0₂₂/D0₂₃
intermetallics: Understanding the stability of silicon substitution

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Synthesis and characterization of (Al,Si)₃(Zr,Ti)-D0₂₂/D0₂₃ intermetallics: Understanding the stability of silicon substitution

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Electronic Supplementary Material (ESM)

1. CRYSTAL SITES OF THE D0₂₂ AND D0₂₃ STRUCTURES

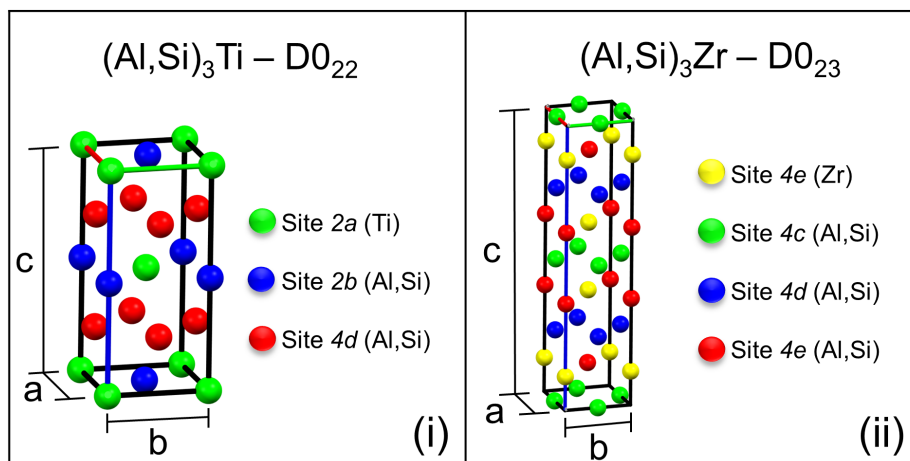


Figure E1. Schematic distribution of crystal sites in $(\text{Al,Si})_3\text{Ti}-\text{D0}_{22}$ (i) and $(\text{Al,Si})_3\text{Zr}-\text{D0}_{23}$ (ii) structures.

2. SIMULATED TEM DIFFRACTION PATTERNS

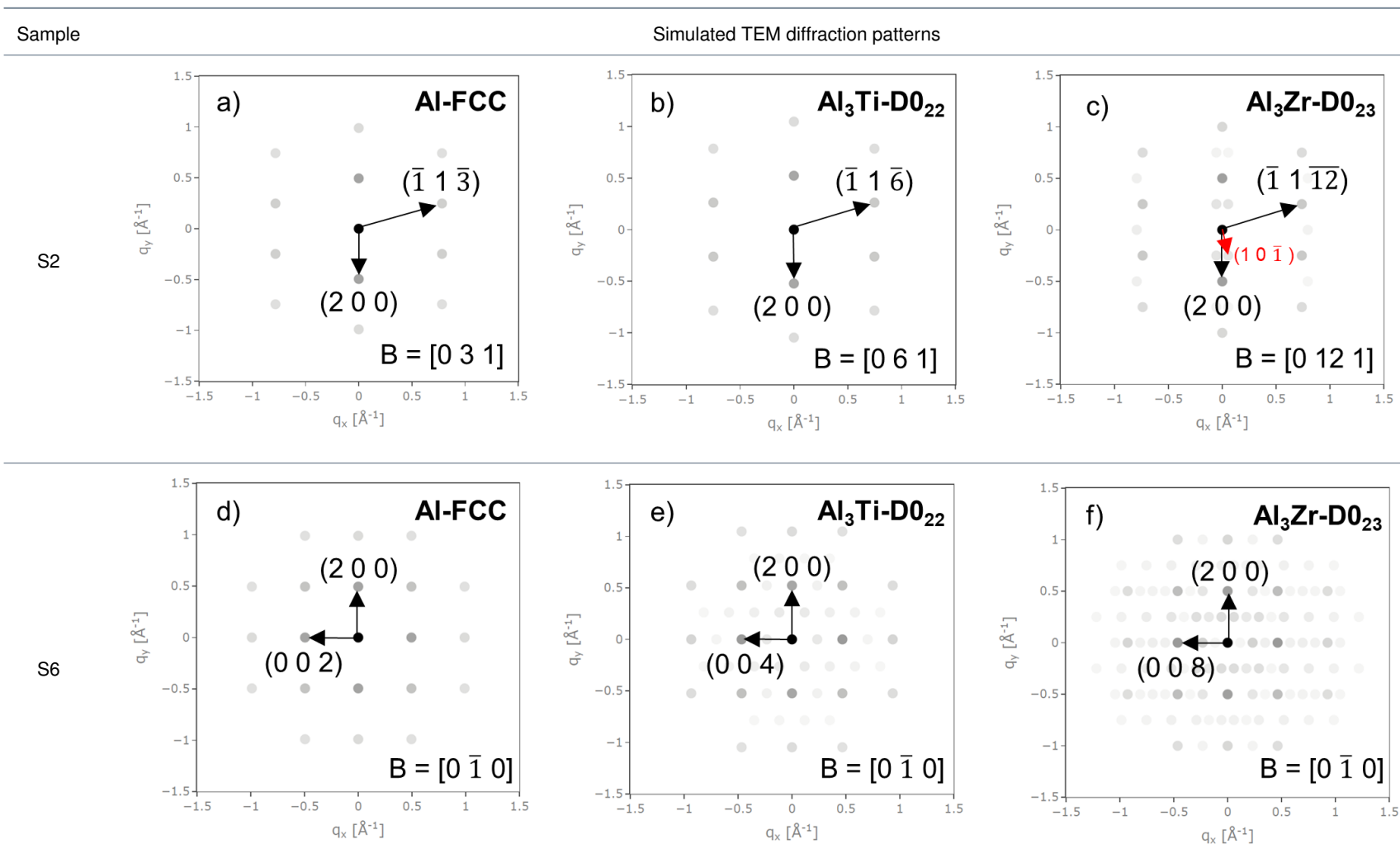


Table E1. Simulated electron diffraction patterns (single crystal) of Al-FCC; Al₃Ti-D0₂₂ and Al₃Zr-D0₂₃ phases. Patterns were computed using the Diffraction module of the Materials Project (MP) database [1]. Beam directions are equivalent to the TEM experimental measurements of this work. A voltage of 200 kV and a maximum scattering angle of 1.25 Å were specified. (a), (b) and (c) patterns correspond to the conditions of sample S2. (d), (e), and (f) are comparable to experimental TEM measurements of sample S6.

3. FORMATION ENTHALPY OF END-MEMBERS IN D0₂₂ AND D0₂₃ SOLID SOLUTIONS

Table E2. Total energy (E_{tot}) and formation enthalpy (ΔH_f) of the $(Al,Si)_3Ti$ -D0₂₂ and $(Al,Si)_3Zr$ -D0₂₂ end-member. Reference states are Al (*A1-fcc*), Si (*A4-diamond*), Ti (*A3-hcp*) and Zr (*A3-hcp*) pure elements.

Crystal sites			E_{tot}	ΔH_f
2a	2b	4d	eV	kJ/mol-at
Ti	Al	Al	-41.322	-38.771
Ti	Si	Al	-44.619	-43.959
Ti	Al	Si	-46.715	-34.685
Ti	Si	Si	-49.095	-28.823
Zr	Al	Al	-43.196	-44.329
Zr	Si	Al	-46.456	-49.082
Zr	Al	Si	-48.487	-39.013
Zr	Si	Si	-51.026	-35.070

Table E3. Total energy (E_{tot}) and formation enthalpy (ΔH_f) of the D0₂₃ end-member. Reference states are Al (*A1-fcc*), Si (*A4-diamond*), Zr (*A3-hcp*) and Ti (*A3-hcp*) pure elements.

Crystal sites				E_{tot}	ΔH_f
4c	4d	4e	4e	eV	kJ/mol-at
Al	Al	Al	Zr	-86.841	-47.033
Al	Al	Si	Zr	-92.460	-46.353
Si	Al	Al	Zr	-92.239	-45.021
Al	Si	Al	Zr	-92.040	-43.822
Si	Al	Si	Zr	-96.984	-39.072
Al	Si	Si	Zr	-97.763	-43.773
Si	Si	Al	Zr	-96.777	-37.821
Si	Si	Si	Zr	-101.877	-34.013
Al	Al	Al	Ti	-82.721	-39.230
Al	Al	Si	Ti	-88.572	-39.949
Si	Al	Al	Ti	-88.245	-37.977
Al	Si	Al	Ti	-88.170	-37.525
Si	Al	Si	Ti	-93.080	-32.569
Al	Si	Si	Ti	-93.778	-36.778
Si	Si	Al	Ti	-93.104	-32.714
Si	Si	Si	Ti	-97.779	-26.341

4. SITE OCCUPANCY OF METASTABLE END-MEMBERS

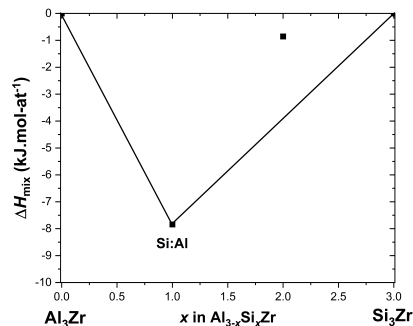


Figure E2. Mixing enthalpy of $(Al,Si)_3Zr$ -D0₂₂ solid solution calculated by DFT at 0 K. The ground-state end-member is labeled with the first element occupying the site 2b, the second element occupying the site 4d. The ground-state is represented by the solid black line.

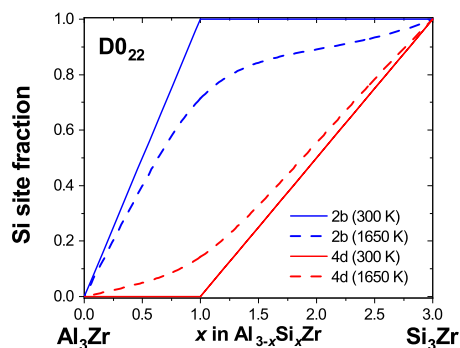


Figure E3. Site fraction of Si on sites 2b and 4d of the D0₂₂ structure calculated at 300 K (solid line) and 1650 K (dashed line).

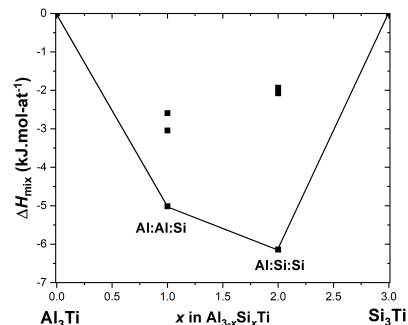


Figure E4. Mixing enthalpy of $(Al,Si)_3Ti$ -D0₂₃ solid solution calculated by DFT at 0 K. The ground-state end-member is labeled with the first, second, and third elements occupying sites 4c, 4d and 4e, respectively. The solid black line represents the ground state.

REFERENCES

1. A. Jain, S. P. Ong, G. Hautier, W. Chen, W. D. Richards, S. Dacek, S. Cholia, D. Gunter, D. Skinner, G. Ceder *et al.*, "Commentary: The materials project: A materials genome approach to accelerating materials innovation," *APL materials* **1**, 011002 (2013).