UF | UNIVERSITY of FLORIDA

Lightweight High Temperature Alloys based on the AlFeSi System

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University of Florida

June 20, 2018

Project ID: MAT069

Overview

Timeline

- Start Data: October 01, 2016
- End Data: December 31, 2019
- Percent Complete: 40%

Budget

- Total Project Cost: $1,102,082
  - DOE share: $991,873
  - Non-DOE share: $110,209
- Funding received in FY17: $342,611
- Funding for FY18: $320,194

Project partners

GM

Oak Ridge National Laboratory

Barriers

- **Weight:** Light-weight materials are needed for vehicle weight reduction in order to meet the future more stringent fuel economy standards
- **Cost:** Low-cost aluminum alloys with high-temperature properties are not available
- **Fabrication:** The τ_{10}-$Al_4Fe_{1.7}Si$ phase has an extremely small composition range and a small fluctuation in composition will change the solidification path creating an unwanted microstructure

Relevance/Objectives

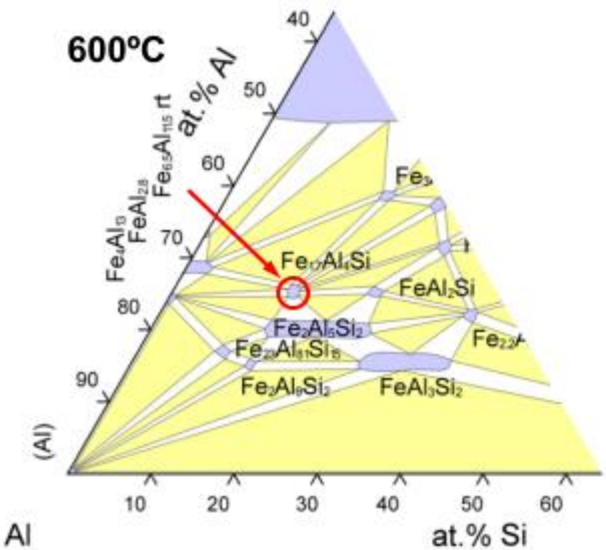
Project Objective:

- To develop low-cost lightweight τ_{10}-$Al_4Fe_{1.7}Si$ based alloys that meet or exceed the high-temperature performance of more expensive Ni-based superalloys and titanium aluminides by combining alloy chemistry strategies and 3D additive manufacturing technologies

Relevance to Weight, Cost & Performance barriers:

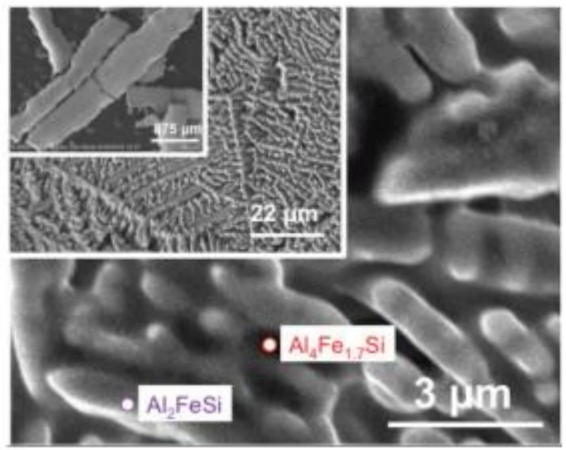
- The high strength-to-weight ratio of τ_{10} makes it ideal for high temperature, lightweight applications
- All the three base constituents are available, abundant in nature, and low cost
- Additive manufacturing enable high cooling rate and can produce components to exact shape with no additional machining

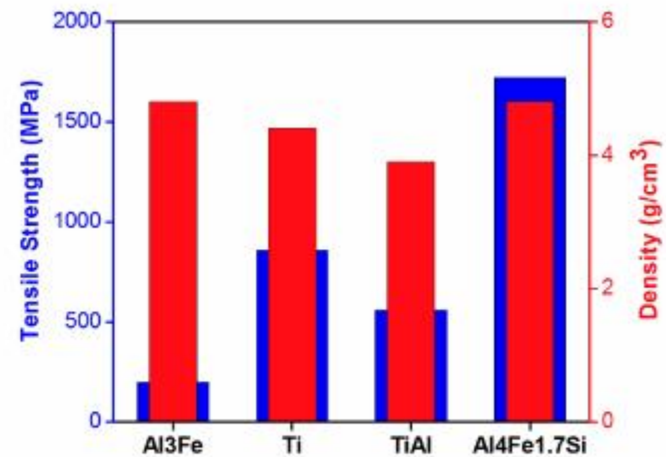
Relevance



ASM Phase Diagram Center

τ_{10}-$Al_4Fe_{1.7}Si$ Phase





General Motors (GM)

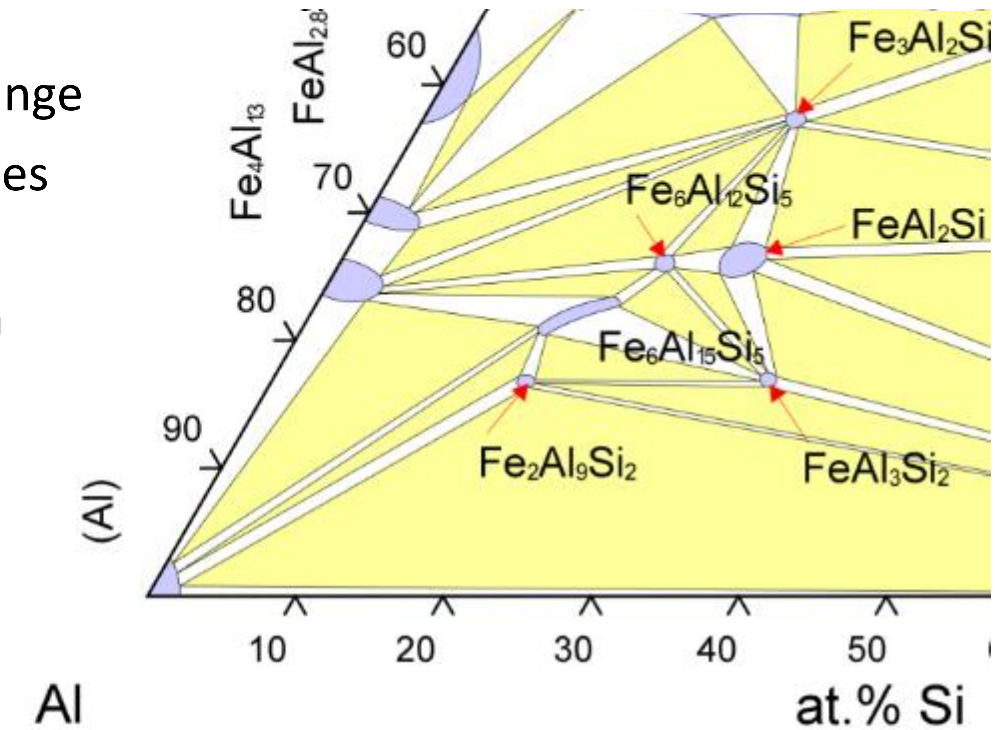
Exceed the room temperature specific strengths of Al_3Fe, Ti and titanium aluminides

Very high strength/weight and thermal stability but little research

Relevance

- Extremely limited composition range
- Large number of competing phases at high temperature
- No longer a stable phase at room temperature (25 °C)

25°C



ASM Phase Diagram Center

Needs either:

- Phase stabilization
- Non-equilibrium solidification

Approaches:

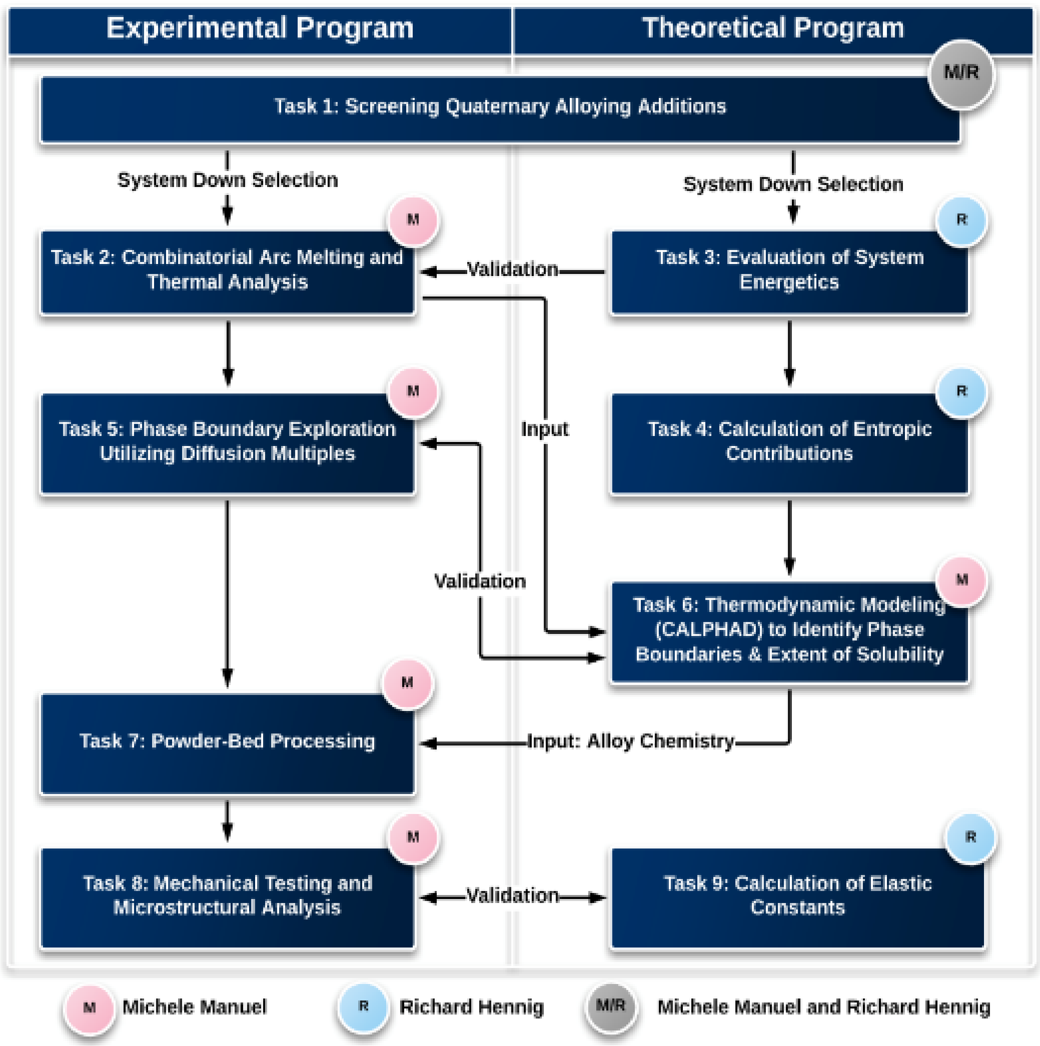
- Expansion of the composition range by addition of minor alloying elements
- High cooling rate enabled by 3D laser printing

Milestones

Budget	Milestone	Planned Completion	Actual Completion	Percent Completion
Period 1	**Quaternary systems down-selected:** Identify potential quaternary solute elements that increase the composition range	Oct-17	Oct-17	100%
	Quaternary systems fabrication complete: Creation of arc melted buttons that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%
	Initial alloys delivered: Deliver alloys that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%
Period 2	**Database of compound thermodynamics developed:** Calculation and evaluation of solute components that stabilize the phase of interest while destabilizing competing phases	Oct-18		15%
	Solution modeling of phase stability ranges at high and low temperatures: Experimentally-validated thermodynamic models will be created that explain each the high temperature solubility and low temperature metastability in quaternary systems of interest	Oct-18		20%
	Chemistry list drafted: Creation of a list of chemistries suitable for powder processing via 3D printing	Oct-18		10%
Period 3	**Production of powders:** Production of powders in the quaternary systems of interest	Jun-19		
	Production of additively manufactured components: Production of mechanical test samples using additive manufacturing	Jun-19		
	Calculation of elastic constants: Database of elastic constants for systems of interest completed	Jun-19		
	Chemistry list completed: List of chemistries are suitable for powder processing via 3D printing completed	Jun-19		

Approach/Strategy

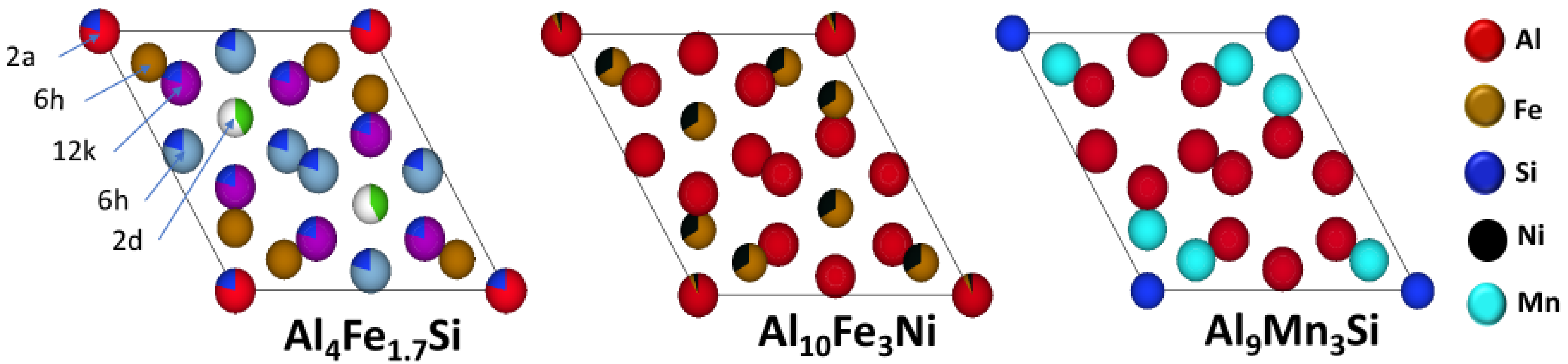
- The overall objective of this project is to combine alloy chemistry and 3D laser printing to create phases with unusual properties
- Scope includes computational and experimental work, culminating in producing 3D laser sintered components for testing



Technical Accomplishments and Progress

Task 1.0 – Screening of quaternary alloying additions

A sorted list of elements that enhances the stability of the τ_{10} phase



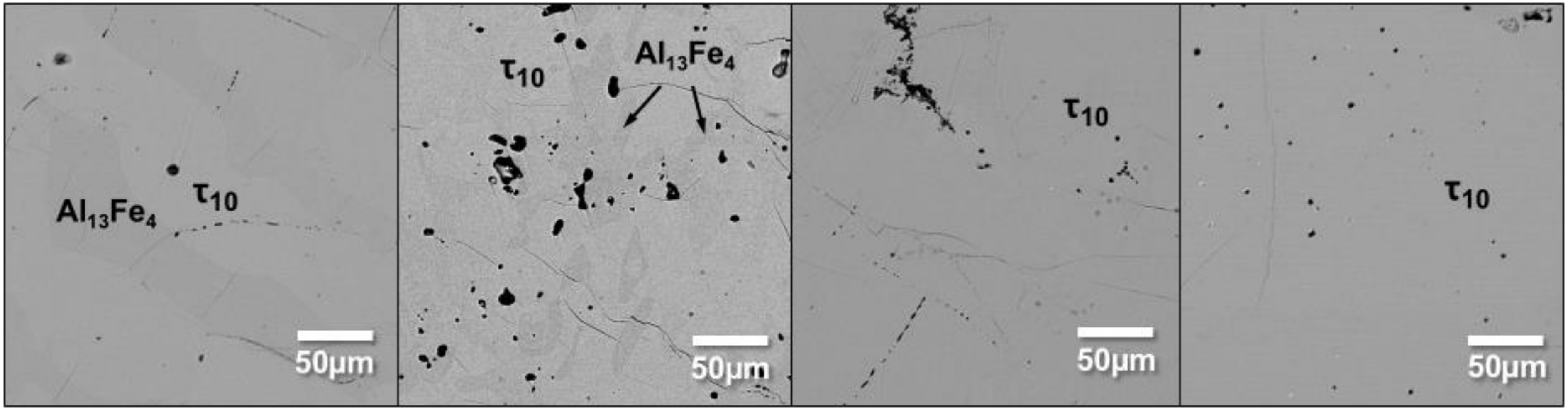
- **Inorganic Crystal Structure Database (ICSD) screening**
- **$Al_{10}Fe_3Ni$ and Al_9Mn_3Si have the similar crystal structure to τ_{10}-$Al_4Fe_{1.7}Si$ (*$P6_3/mmc$*)**
- **Ni and Mn were selected as promising quaternary additions**

Technical Accomplishments and Progress

Task 2.0 – Combinatorial arc melting and thermal analysis

Exploring the stable compositional range of τ_{10}-$Al_4Fe_{1.7}Si$

800 °C, 550 hrs

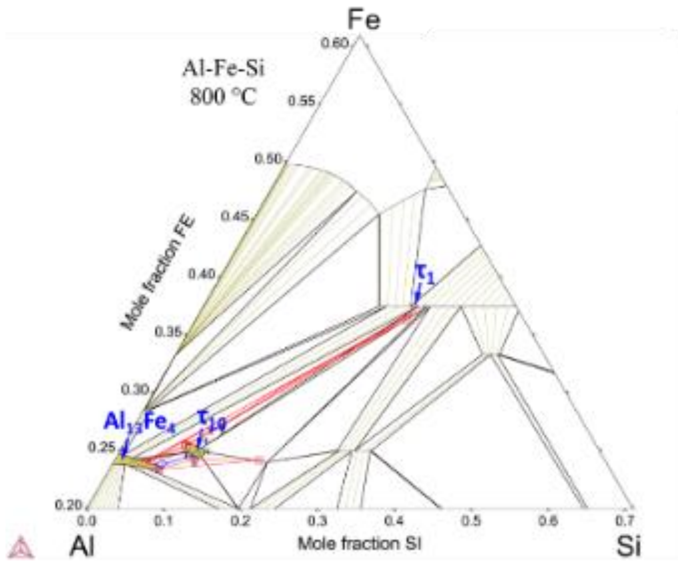


Al-24.5Fe-9.0Si | Al-24.5Fe-10.2Si | Al-24.5Fe-11.0Si | Al-24.5Fe-12.0Si

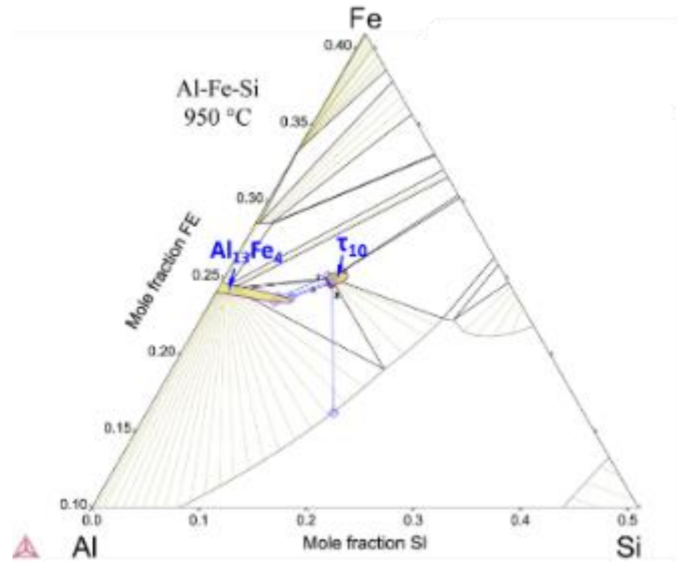
Increasing in Si content (in at.%)

Technical Accomplishments and Progress

Task 2.0 – Combinatorial arc melting and thermal analysis



Al (24.3-25.5)Fe (8.2-10.8)Si



Al (24.6-25.2)Fe (9.6-11.0)Si

Measured stable compositional range of τ_{10}-$Al_4Fe_{1.7}Si$ at 800 & 950 °C

Tasks 1 & 2 Milestone	Planned Completion	Actual Completion	Percent Completion
Quaternary systems down-selected: Identify potential quaternary solute elements that increase the composition range	Oct-17	Oct-17	100%
Quaternary systems fabrication complete: Creation of arc melted buttons that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%
Initial alloys delivered: Deliver alloys that have the potential to display a stable τ_{10}-Al-Fe-Si-X phase	Oct-17	Dec-17	100%

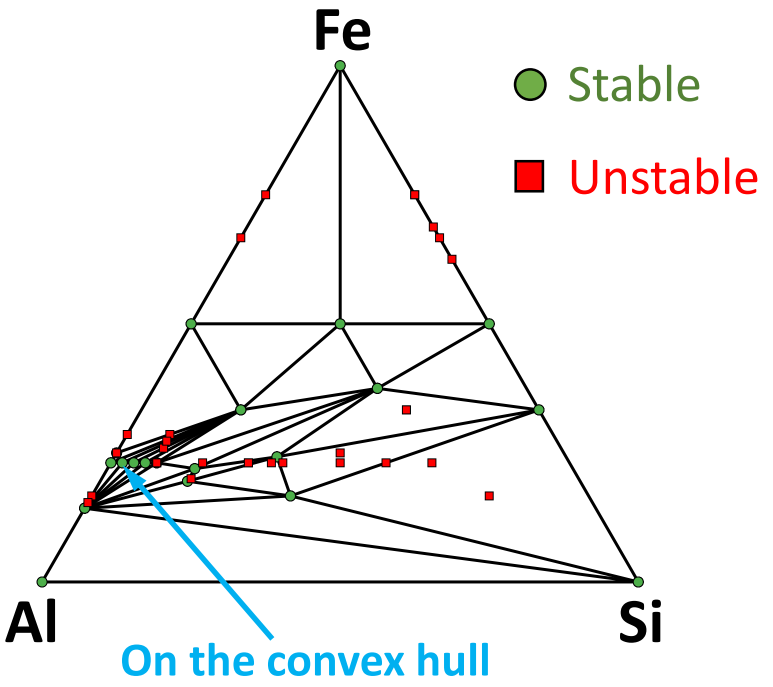
Technical Accomplishments and Progress

Tasks 3.0 & 4.0 – Calculations of system energetics and entropic contributions

Gibbs free energy for the τ_{10} and completing Al-Fe-Si-X phases

Energy Calculations

- Used cluster expansion to generate various configurations
- Structures from Materials project and ICSD database
- Energy calculations with partial occupancies consideration
- Convex hull construction

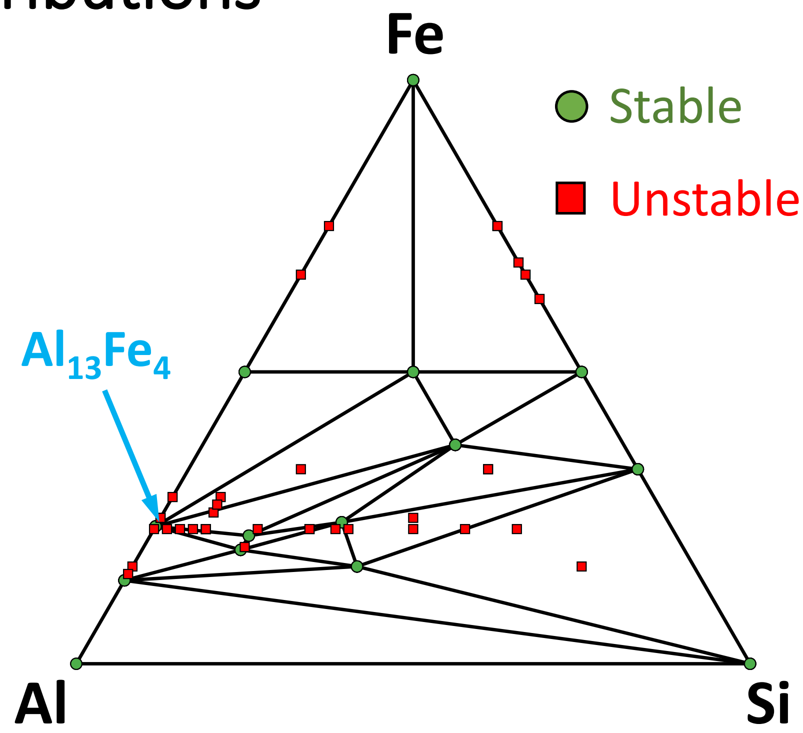


Technical Accomplishments and Progress

Tasks 3.0 & 4.0 – Calculations of system energetics and entropic contributions

Energy Calculations

- Experimentally, $Al_{13}Fe_4$ phase along with τ_{10} in AlFeSi and AlFeNi systems
- Including $Al_{13}Fe_4$ in calculations pulled the convex hull down

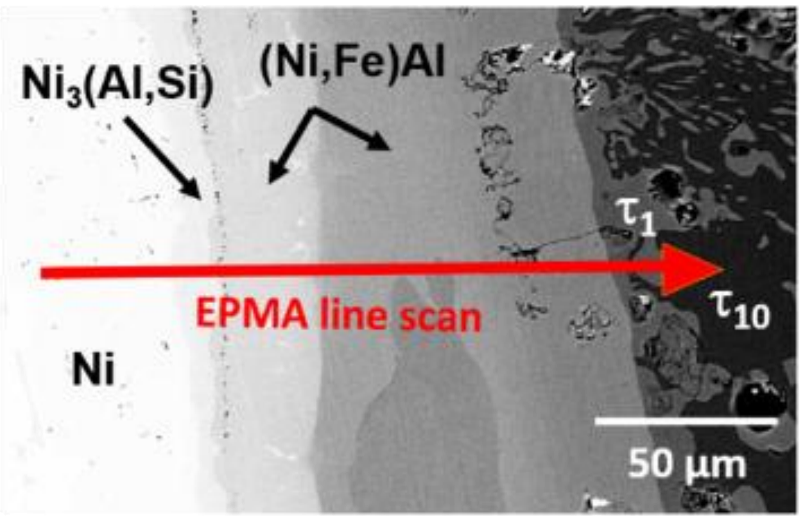


Tasks 3 & 4 Milestone	Planned Completion	Actual Completion	Percent Completion
Database of compound thermodynamics developed: Calculation and evaluation of solute components that stabilize the phase of interest while destabilizing competing phases	Oct-18		20%

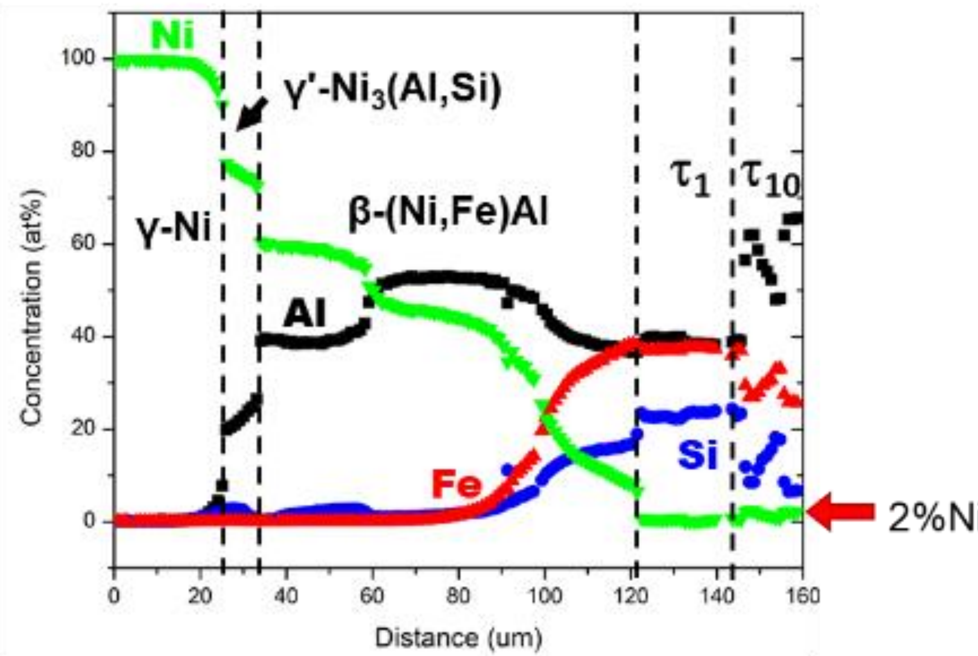
Technical Accomplishments and Progress

Task 5.0 – Phase boundary exploration: diffusion couple

Exact solubility limit of the quaternary solutes in the τ_{10} phase



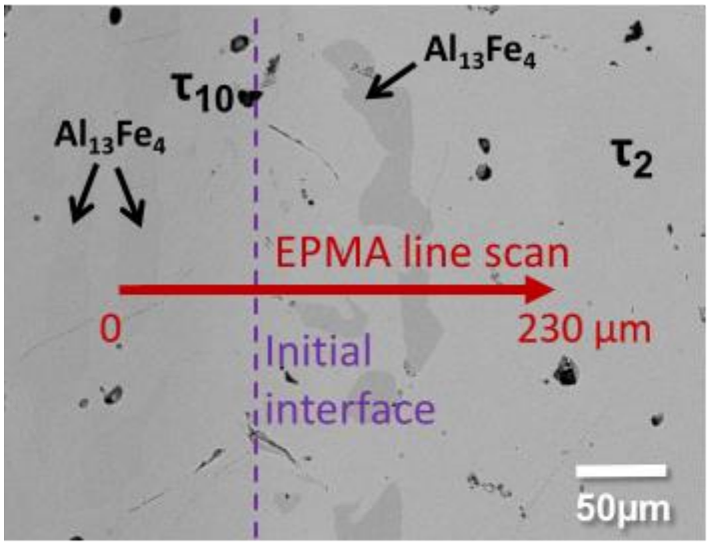
Ni/Al-24.5Fe-10.2Si diffusion couple
800 °C, 336 h



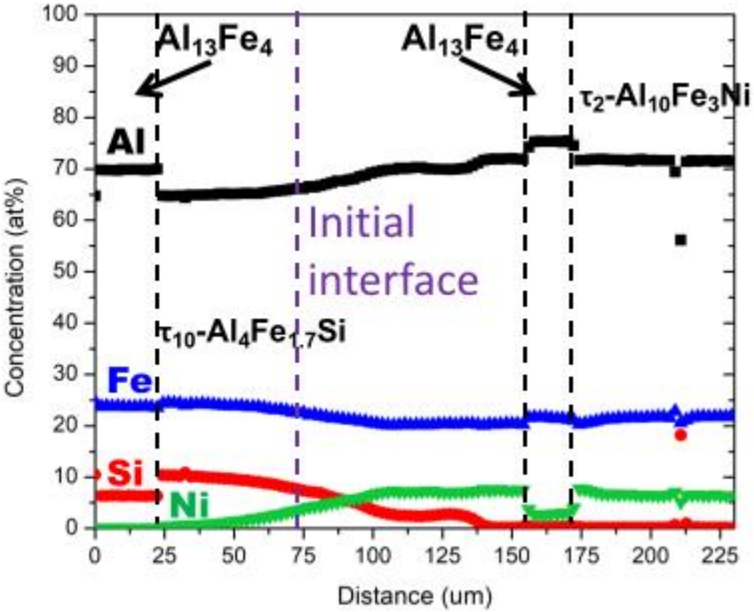
Solubility of Ni in τ_{10}-$Al_4Fe_{1.7}Si$ at 800°C was determined to be 2 at.%

Technical Accomplishments and Progress

Task 5.0 – Phase boundary exploration: diffusion couple



Al-24.5Fe-10.2Si / Al-21.5Fe-7.0Ni diffusion couple
800 °C, 336 h

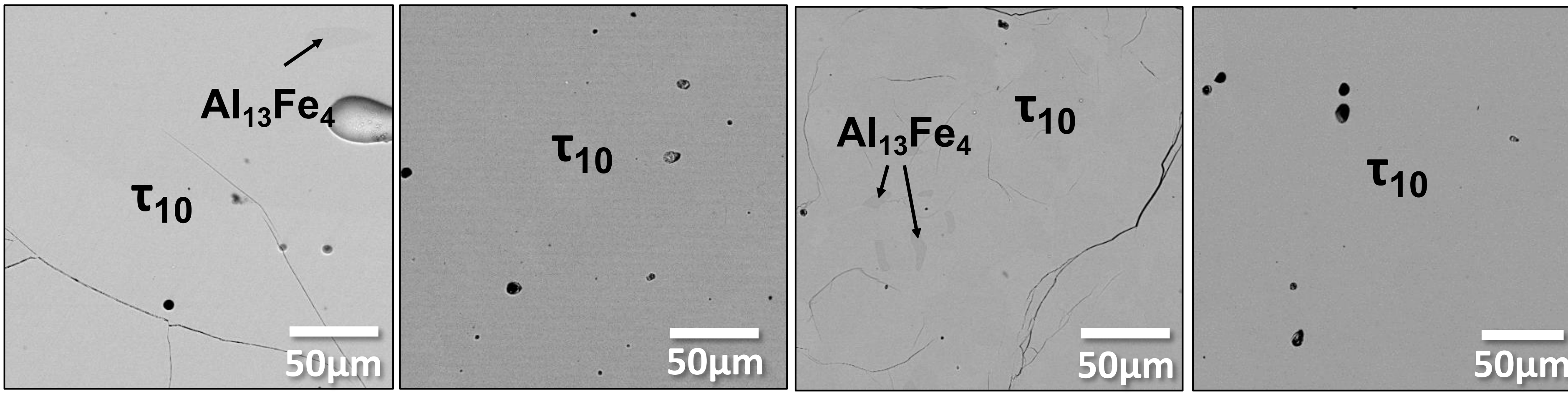


Ni predominately replaces Si while the Al, Fe contents in the τ_{10} phase changes only slightly

Technical Accomplishments and Progress

Task 5.0 – Phase boundary exploration: alloy verification

900 °C, 150 hrs



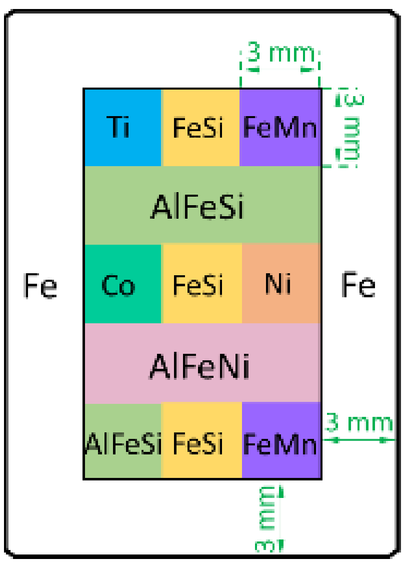
Al-24.5Fe-9.2Si-1.0Ni | **Al-24.5Fe-8.2Si-2.0Ni** | **Al-25.7Fe-7.7Si-1.0Ni** | **Al-25.7Fe-6.7Si-2.0Ni**

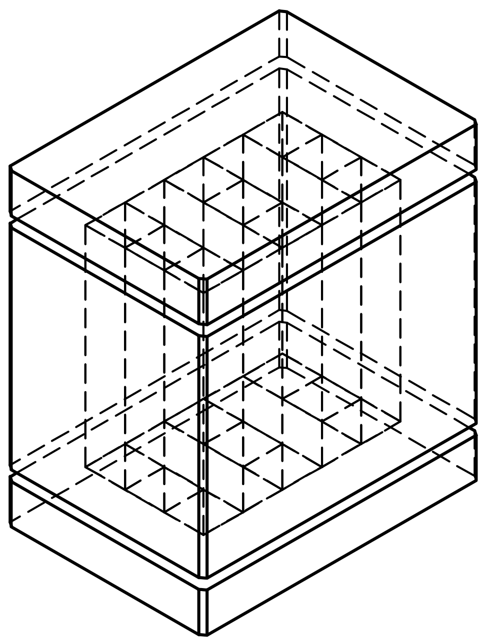
Alloys with 2 at.% Ni were confirmed to be the τ_{10}–Al-Fe-(Si, Ni) single phase

Technical Accomplishments and Progress

Task 5.0 – Phase boundary exploration: diffusion multiple

Al-Fe-Si-X (X = Co, Mn, Ni, Ti) diffusion multiples to accelerate the measurement of solubility limits of quaternary solutes in τ_{10}





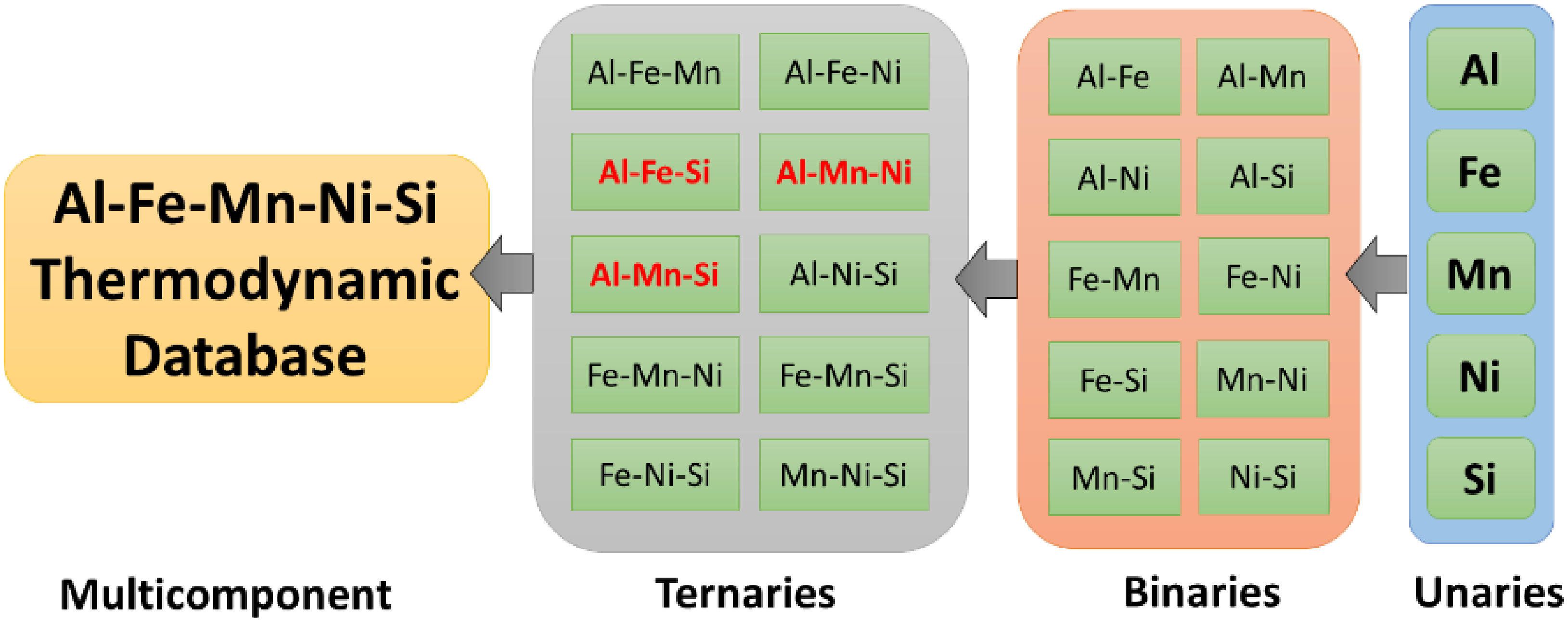
16 Quaternary Systems:

AlFeSi/Fe/Co	AlFeSi/FeSi/Co
AlFeSi/Fe/FeMn	AlFeSi/FeSi/FeMn
AlFeSi/Fe/Ni	AlFeSi/FeSi/Ni
AlFeSi/Fe/Ti	AlFeSi/FeSi/Ti
AlFeNi/Fe/Co	AlFeNi/FeSi/Co
AlFeNi/Fe/FeMn	AlFeNi/FeSi/FeMn
AlFeNi/Fe/Ni	AlFeNi/FeSi/Ni
AlFeNi/Fe/AlFeSi	AlFeNi/FeSi/AlFeSi

Technical Accomplishments and Progress

Task 6.0 – Thermodynamic Modeling

Update the parameters for the Al-Fe-Si, Al-Mn-Ni and Al-Mn-Si ternary systems



Tasks 5 & 6 Milestone	Planned Completion	Actual Completion	Percent Completion
Solution modeling of phase stability ranges at high and low temperatures: Experimentally-validated thermodynamic models will be created that explain each the high temperature solubility and low temperature metastability in quaternary systems of interest	Oct-18		20%

Responses to Previous Year Reviewers' Comments

No previous year reviewers' comments are available

Partnerships and Collaborations

GM

Provided induction melted ingots and has participated in monthly and annual onsite meetings. The continually offer advice and recommendations on solute selection and alloy design

They will next be producing powders of our AlFeSiX alloy in late 2018 for additive manufacturing

Oak Ridge National Laboratory

Worked with beamline scientist to study site-lattice occupancy of elements within the τ_{10}-$Al_4Fe_{1.7}Si$. This information will be used to validate computational models

Remaining Challenges and Barriers

- The τ_{10}-$Al_4Fe_{1.7}Si$ phase is a high temperature phase which is only stable between 727 and 997 °C. The high cooling rate (~10^7 K/s - ~10^{13} K/s) enabled by 3D laser printing to selectively promote the solidification of τ_{10} single phase. Therefore, the proper processing conditions for 3D laser printing need to be explored to control the solidification path without creating an unwanted microstructure
- High thermodynamic stability of competing phase add an extra layer of complexity as additional solute elements should stabilize the τ_{10}-$Al_4Fe_{1.7}Si$ phase while de-stabilizing detrimental competing phases such as $Al_{13}Fe_4$
- High number of atoms in the system adds complexity to the cluster expansion simulations
- Partial occupancies present in the system a large number of potential configurations

Future work

- ❑ Continue to expand the phase boundaries of the τ_{10}-$Al_4Fe_{1.7}Si$ intermetallic phase with the addition of minor quaternary alloying elements
- ❑ Provide phase stability maps for the τ_{10} and completing phases in the Al-Fe-Si-X system to inform sample preparation for 3D printing and mechanical testing
- ❑ Obtain a list of AlFeSiX chemistries that are suitable for powder processing via 3D printing and have excellent high-temperature performance

Ongoing	**FY 2018**	**Q4 Milestone (Tasks 3 & 4) — Database of compound thermodynamics developed:** Calculation and evaluation of solute components that stabilize the phase of interest while destabilizing competing phases
		Q4 Milestone (Tasks 5 & 6) — Solution modeling of phase stability ranges at high and low temperatures: Experimentally-validated thermodynamic models will be created that explain each the high temperature solubility and low temperature metastability in quaternary systems of interest
		Q4 Go/No Go Decision Point — Chemistry list drafted: Creation of a list of chemistries suitable for powder processing via 3D printing
Proposed	**FY 2019**	**Q4 Milestone (Task 7) — Production of powders:** Production of powders in the quaternary systems of interest
		Q4 Milestone (Task 8) — Production of additively manufactured components: Production of mechanical test samples using additive manufacturing
		Q4 Milestone (Task 9) — Calculation of elastic constants: Database of elastic constants for systems of interest completed
		Q4 Go/No Go Decision Point —Chemistry list completed: List of chemistries are suitable for powder processing via 3D printing completed

Any proposed future work is subject to change based on funding levels

Summary

Project Objective:

- To develop low-cost lightweight τ_{10}-$Al_4Fe_{1.7}Si$ based alloys that meet or exceed the high-temperature performance of more expensive Ni-based superalloys and titanium aluminides by combining alloy chemistry strategies and 3D additive manufacturing technologies

Results:

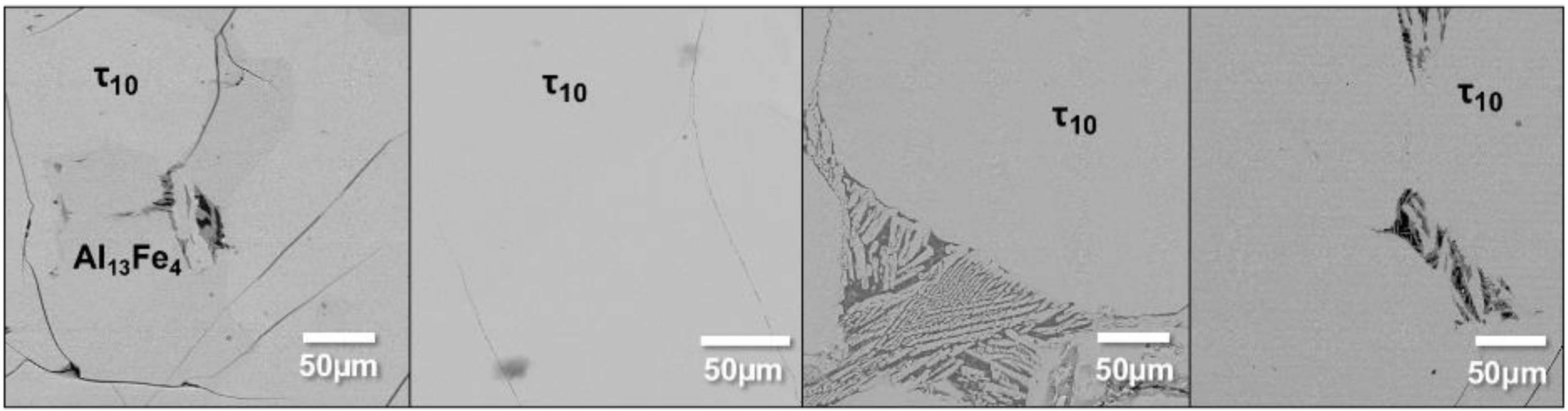
- $Al_{10}Fe_3Ni$ and Al_9Mn_3Si have the similar crystal structure to τ_{10}-$Al_4Fe_{1.7}Si$ (*P63/mmc*)
- Co, Cu, Mn, Ni and Ti were selected as promising quaternary additions to expand the composition range of τ_{10}-$Al_4Fe_{1.7}Si$
- Regular solution model was used to determine the configurational entropy and included to the energy calculations. The data from regular solution model was compared to the data from the Monte Carlo simulations. This has been done for the Nb-Sn binary system as the concept of proof for the method
- The stable compositional range of the τ_{10}-$Al_4Fe_{1.7}Si$ at 800 °C and 950 °C was determined to be Al-(24.3-25.5)Fe-(8.2-10.8)Si and Al-(24.6-25.2)Fe-(9.6-11.0)Si, respectively
- The solubility of Ni in τ_{10}-$Al_4Fe_{1.7}Si$ at 800 °C and 850 °C was determined to be 2 at.% and 2.7 at.%, respectively, predominately replacing Si
- Two quaternary alloys with 2 at.% Ni were fabricated and confirmed to be the τ_{10}–Al-Fe-(Si, Ni) single phase

Back-up Slides

Task 2.0 – Combinatorial arc melting and thermal analysis

Exploring the stable compositional range of τ_{10}-$Al_4Fe_{1.7}Si$

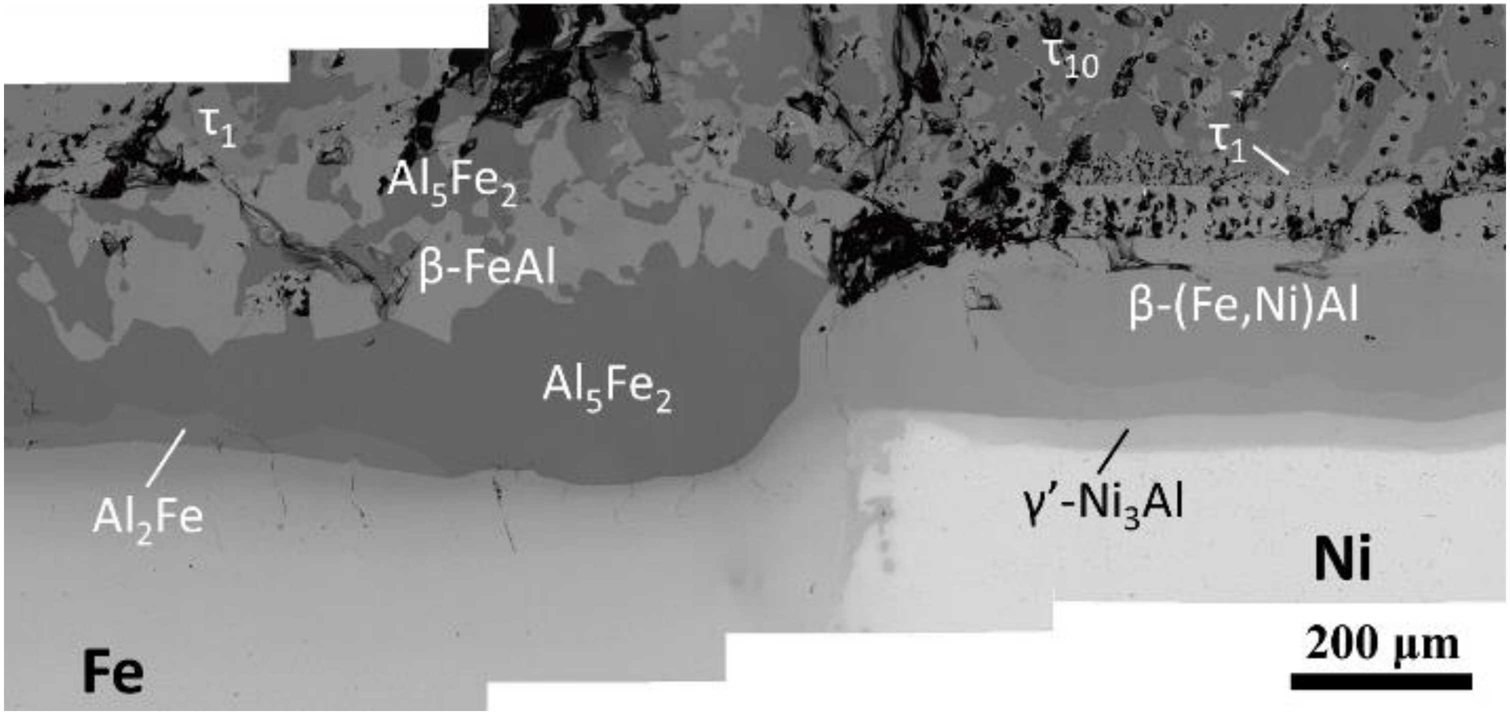
950 °C, 100 hrs



Al-24.5Fe-9.0Si | Al-24.5Fe-10.2Si | Al-24.5Fe-11.0Si | Al-24.5Fe-12.0Si

Increasing in Si content (in at.%)

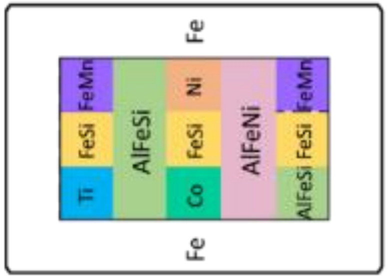
Task 5.0 – Phase boundary exploration: diffusion triple



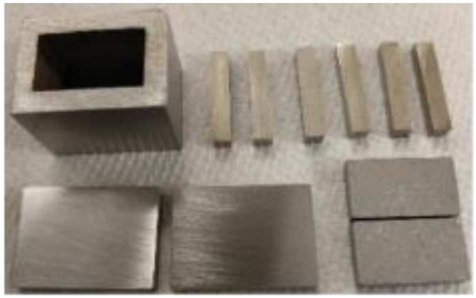
Ni/Fe diffusion couple: 1050 °C, 310 h
Ni/Fe/AlFeSi diffusion triple: 850 °C, 340 h

Solubility of Ni and Fe in τ_{10}-$Al_4Fe_{1.7}Si$ at 850°C was determined to be 2.7 at.% and 27 at.%, respectively

Al-Fe-Si-X Diffusion multiple fabrication



Design

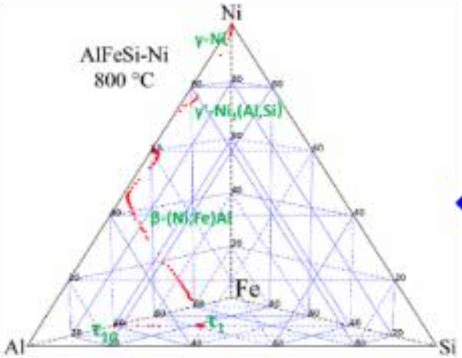


Grind & Assemble

Electron Beam (EB) Welding

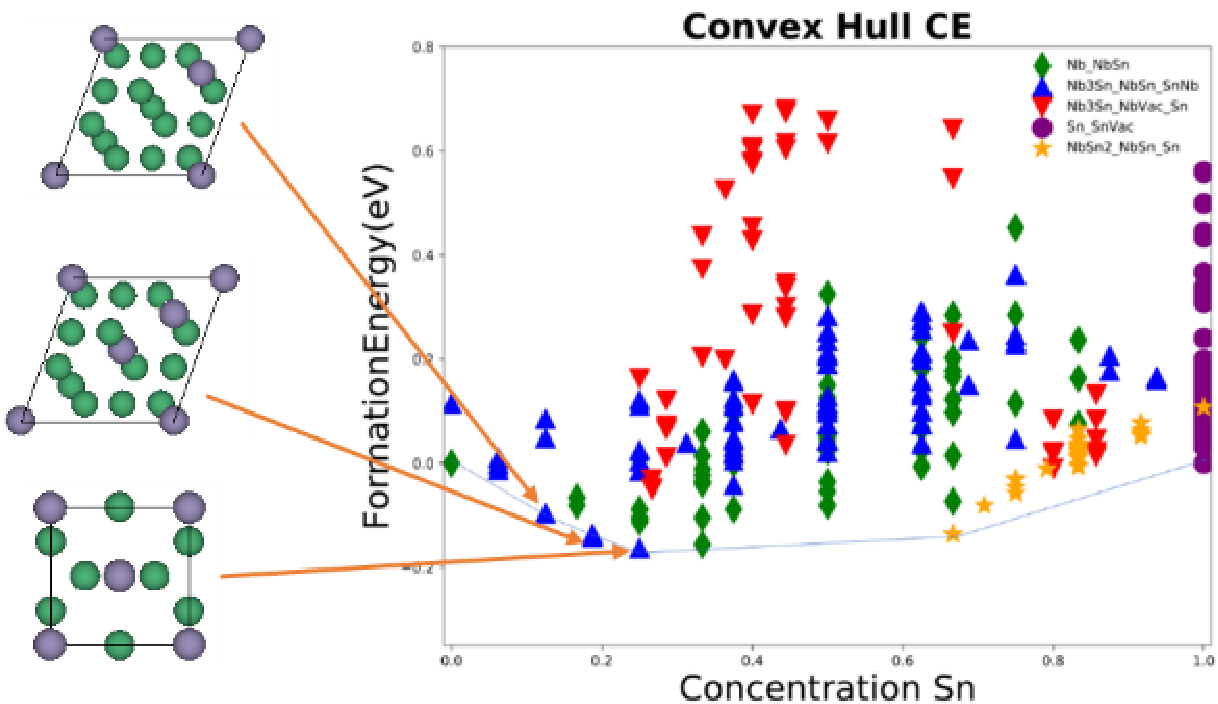
Hot Isostatic Pressing (HIP)

Encapsulation & Annealing



Characterization & Analysis

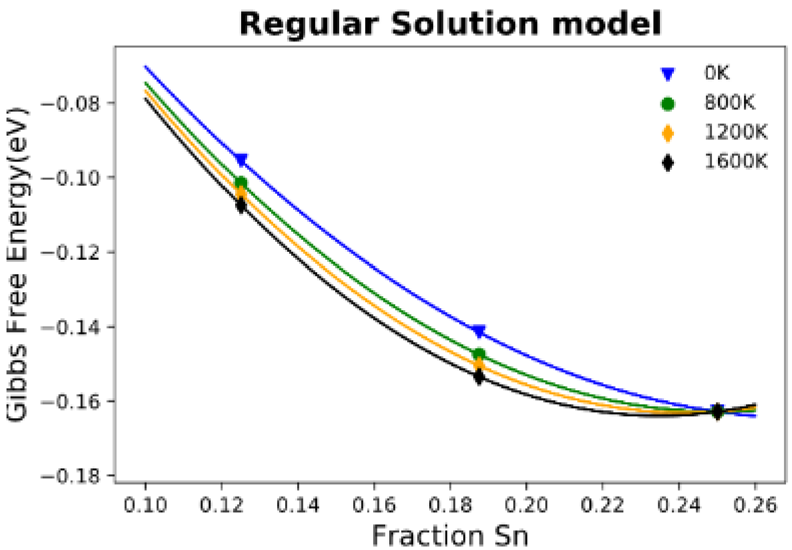
Tasks 3.0 & 4.0 – Calculations of System Energetics and Entropic Contributions

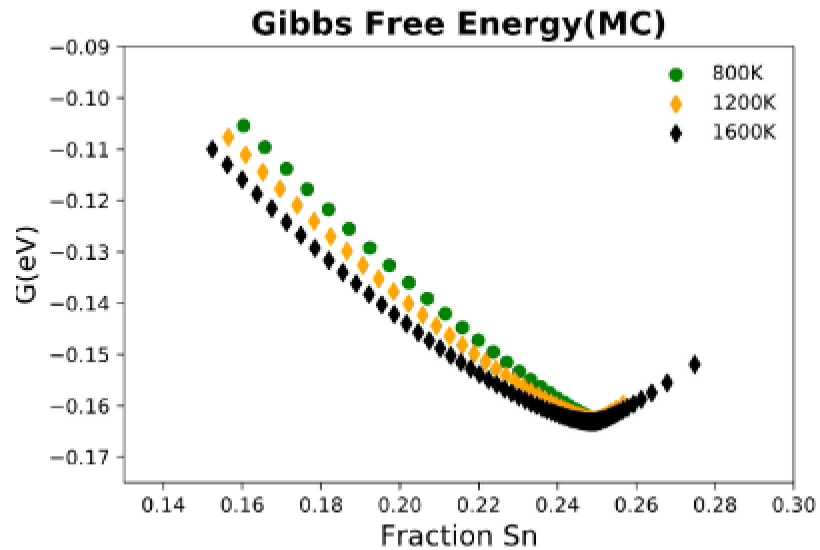


Convex hull in Nb-Sn binary system to show proof of concept

Tasks 3.0 & 4.0 – Calculations of System Energetics and Entropic Contributions

Entropic contribution to energy for the Nb-Sn binary system





$$\Delta G = \Delta H_{mix} - kT \sum X_i ln X_i$$

S_{mix}^{ideal}

Semi Grand Canonical Monte Carlo

$$\emptyset(\beta, \mu) = -\frac{1}{\beta N} \ln(\sum_{i=0}^{n} exp^{(-\beta N(E_i - \mu x_i))})$$