Synthesis of Ag-Sn Phases Predicted with Machine Learning

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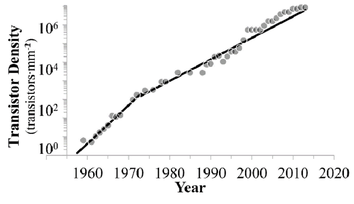
SCALE
SCalable Asymmetric Lifecycle Engagement
Developing the U.S. Defense Microelectronics Workforce

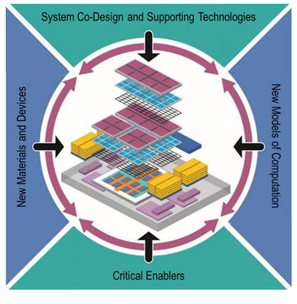
Grand Challenges

- Chip miniaturization limitations
  - Heat flux >350 W/cm^2 impairs cooling in compact systems
  - Joint current density >10^4 A/cm^2 accelerates device failure rates
  - Junction T ≈ 250 °C must be tolerated while resisting thermal and electromigration stress
- Ag-Sn alloys as novel interconnects
  - Up to 4 times greater thermal fatigue resistance vs Sn-Pb alloys
  - Better ductility, EM and creep resistance vs Sn-Pb alloys
  - RoHS-compliant: ≤ 0.1% Pb

Growth of transistor density



Heterogenous integration

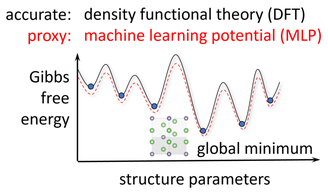


Do any Ag-Sn alloys remain undiscovered?

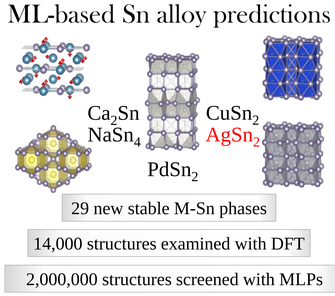
Machine Learning (ML) Predictions

- Standard *ab initio* search strategy (accurate but cost demanding)
  - Define the chemical space
  - Employ global search engine
  - Check T=0 K stability with DFT
- ML screening with MAISE [1,2] (accelerated by a factor of 10^2-10^3)
  - Build MLPs for selected species
  - Perform evolutionary searches
  - Use MLPs to select candidates
  - Check for high-T ground states
  - Check all results with DFT
- Predicted Ag-Sn compounds (brand-new crystal structures shown stable at the DFT level)
  - $AgSn_9$: may form above 360 K
  - $AgSn_4$: may form above 570 K

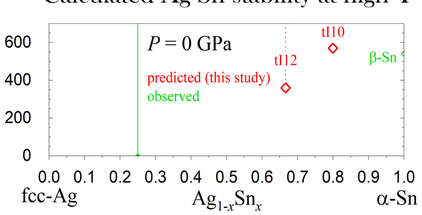
Potential energy surface sampling



ML-based Sn alloy predictions



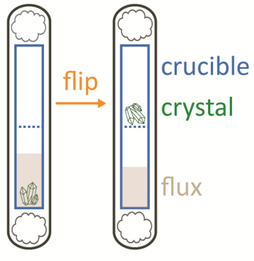
Calculated Ag-Sn stability at high T



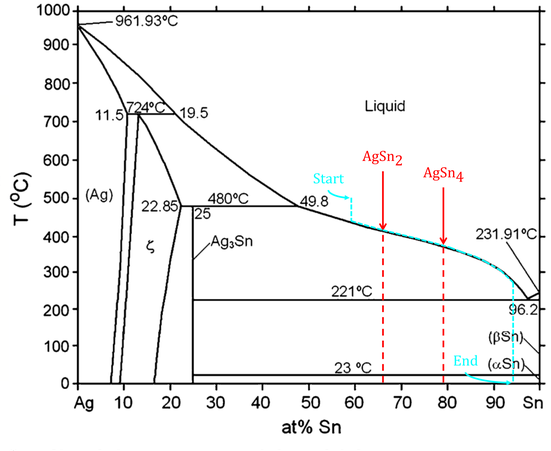
Can the predicted Ag-Sn alloys be synthesized?

Synthesis Strategy

Crystal synthesis via the flux growth method



Ag-Sn binary phase diagram [3]

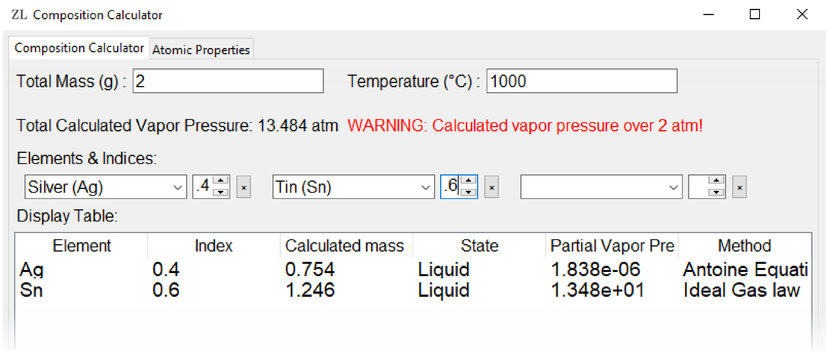


- Tin flux acts as solvent for dissolving raw materials at high temperature
- As the solution slowly cools, the solubility decreases, leading to the gradual crystallization of the Ag-Sn phases

How to ensure the synthesis is safe and efficient?

Vapor Pressure Estimation

Managing the vapor pressure is crucial at high temperatures



We estimate vapor pressure via:

1. Antoine equation (best case)

$$\log_{10}(p) = A - \frac{B}{C+T}$$

2. Ideal gas law (otherwise)

$$PV = nRT$$

p: absolute vapor pressure (mmHg) R: gas constant $\left(0.0821 \frac{L \cdot atm}{mol \cdot K}\right)$

T: temperature (°C) A, B, C: element-specific Antoine coefficients

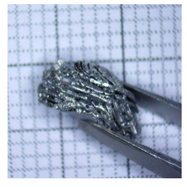
Antoine coefficients and valid temperature values specific to Ag and Sn [4].

	A	B	C	T_{min}	T_{max}
Ag	8.992	1.5884E+4	387.39	960.85	6,136.85
Sn	8.549	1.6656E+4	336.40	1,150.00	2,800.00

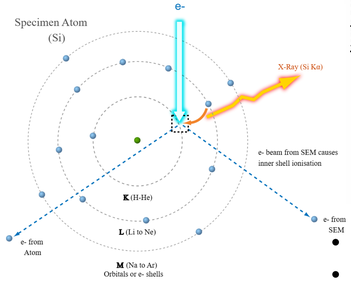
How to compare the synthesis outcome with the prediction?

Preliminary Results

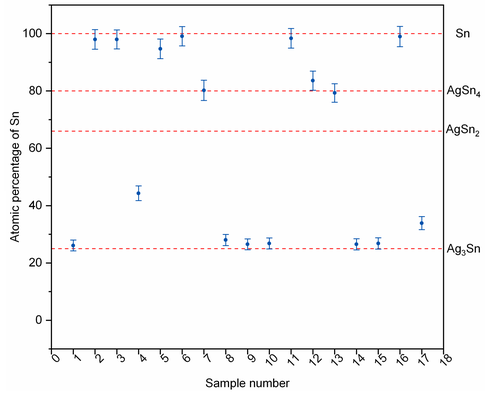
Synthesized Ag-Sn alloy



Element specific X-Ray



Compositional analysis of the synthesized Ag-Sn binary alloy via Energy-Dispersive X-ray spectroscopy.



- $AgSn_4$ is likely present
- $AgSn_2$ is likely present (data not shown here)

Summary and References

Key observations

- Ag-Sn alloys show significant promise as next-generation interconnects
- New Ag-Sn binary phases were predicted by machine learning
- Ag-Sn alloy was synthesized using the flux growth method
- A GUI was developed to estimate the vapor pressure
- Compositional analysis revealed $AgSn_2$ and $AgSn_4$ are likely present

Future work

- Purify the targeted binary phases
- Perform X-ray diffraction to determine the crystal structure
- Test Ag-Sn as interconnects
- Attempt synthesis of other predicted Sn alloys

Primary References

[1] A. Thorn *et al.*, *Physical Chemistry Chemical Physics*, 2023, **25**, 22415

[2] S. Hajinazar *et al.*, *Computer Physics Communications*, 2021, **259**, 107679

[3] J.F. Li, P.A. Agyakwa, and C.M. Johnson, *J. of Electronic Materials*, 2014, **43(4)**, 983

[4] C.L. Yaws, *Chemical Engineering*, 2006, **13 3(12)**, 52

Additional References