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Segregation effects in Sn-Li

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Outline

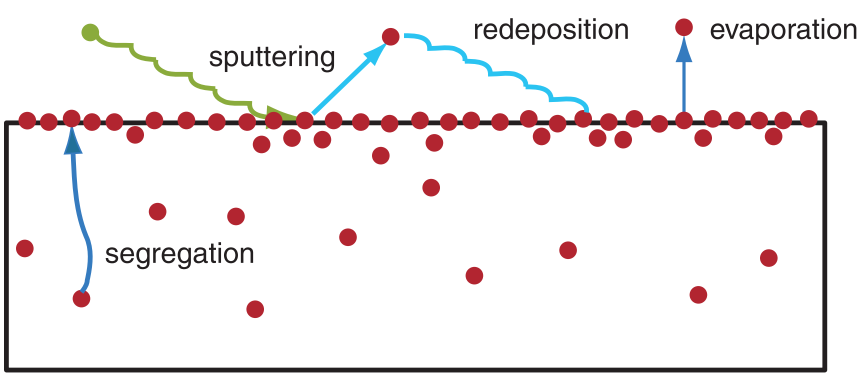
- **Background and motivation**
- **Surface composition data on Sn-Li**
- **Surface model for Sn-Li**
- **Behavior of Sn-Li under plasma conditions**
- **Summary**

Surface studies of plasma-facing liquids

Issue: What comprises the surface seen by the plasma?

Need: Experimental data on liquid surfaces needed to properly model conditions that will exist in a fusion reactor.

Tasks:
- Examine surface composition of candidate liquids
- Consider how surface composition will evolve during plasma exposure due to combined effects of:



> evaporation
> sputtering / redeposition
> segregation.

The surface composition of a liquid can differ from its bulk composition.

- Components that lower surface energy tend to segregate to surfaces.
- For binary liquids, the Gibbsian segregation rule is

$$\gamma_A + \frac{RT}{\sigma_A}\ln\left(\frac{x_s}{x_b}\right) = \gamma_B + \frac{RT}{\sigma_B}\ln\left(\frac{1-x_s}{1-x_b}\right)$$

where

γ_i is surface tension and
σ_i is surface area for species i.

- Example: $\gamma_{\mathrm{Li}} < \gamma_{\mathrm{Sn}}$ (385 vs. 520 erg/cm^2)
  $\Rightarrow$ Li segregates to Sn-Li surfaces.

Refs: J. W. Gibbs, The Scientific Papers of J. Willard Gibbs, Vol. 1 (Longmans, Green, New York, 1906), p. 219.
M. J. Regan, et al., Phys. Rev. B 55 (1997) 15874.

Surface analysis of liquid tin-lithium

- $Sn_{0.8}Li_{0.2}$ was prepared in-situ by melting high-purity Sn and Li foils.
- Low-energy ion scattering spectroscopy was used to measure the surface composition. The He^+ probe also served to sputter clean the liquid surface.
- After melting, the sample was flashed to 800 °C to promote formation of a single-phase binary liquid.
- Equilibrium and transient surface composition measurements were made from the melting point to 500 °C.

→ This enabled segregation behavior to be examined.

In-situ preparation of liquid 80% Sn - 20% Li alloy

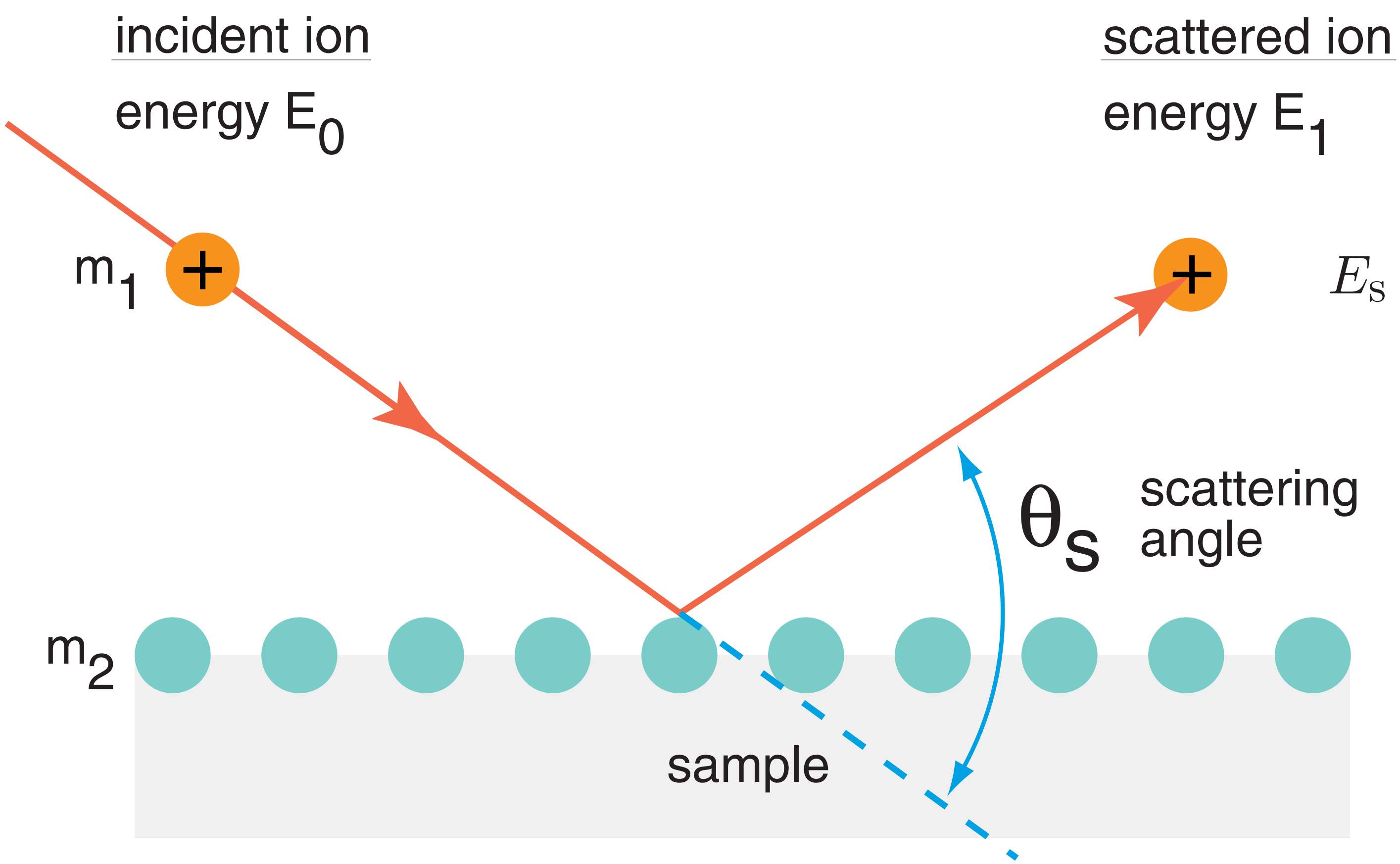
$$\frac{d_{Li}}{d_{Sn}} = \frac{V_m(Li)\ f(Li)}{V_m(Sn)\ f(Sn)}$$

$V_m(Li) = 13.1\ cm^3$

$V_m(Sn) = 16.3\ cm^3$

- Layers of high-purity Sn and Li foils are melted in vacuum.
- A 5:1 thickness ratio produces the desired stoichiometry.
- The top and bottom layers are Sn to reduce Li evaporation.

Surface measurements consist of aiming a monoenergetic ion beam at a surface and measuring the energy loss of reflected ions.

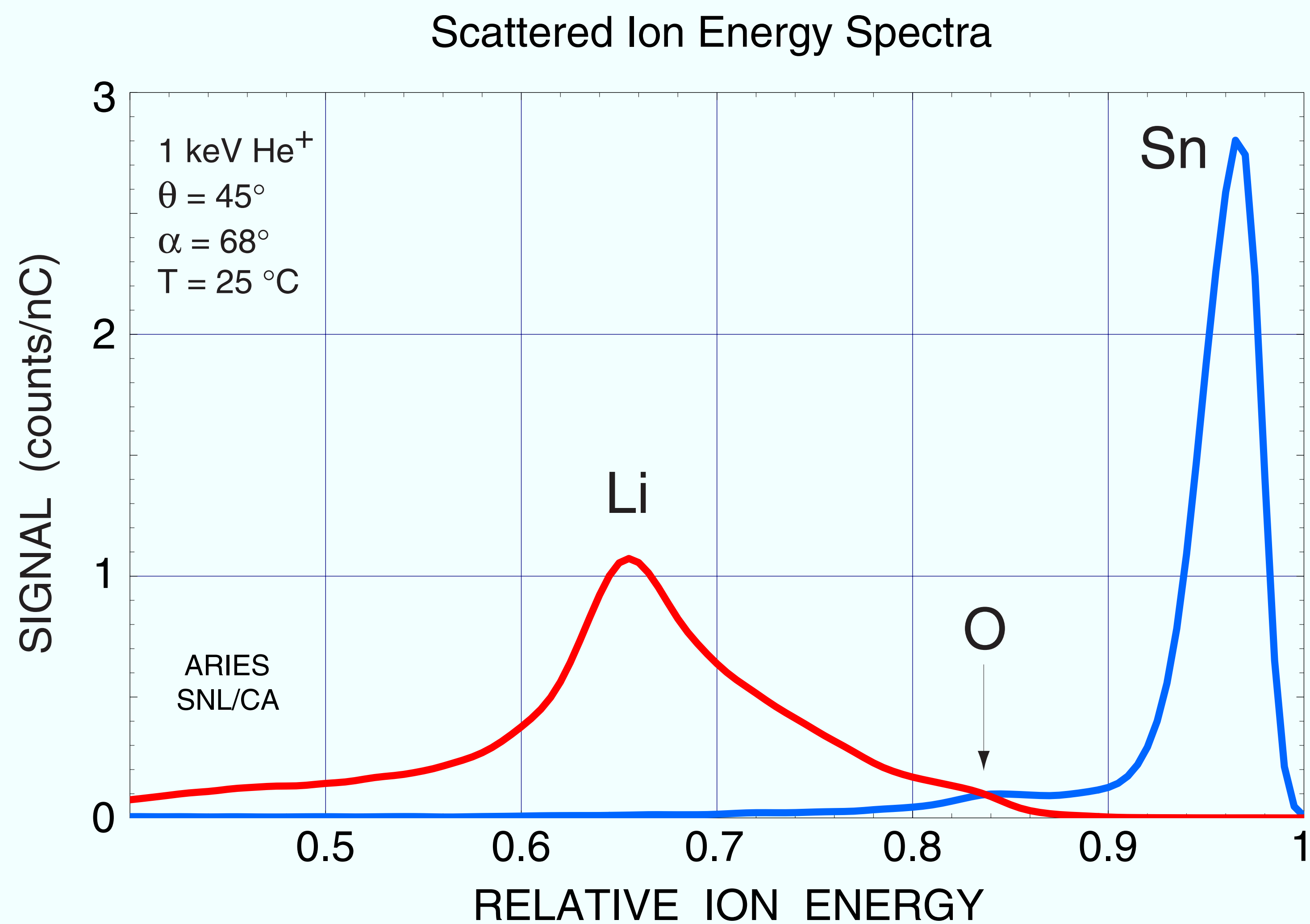


$$E_S = \frac{E_1}{E_0}$$

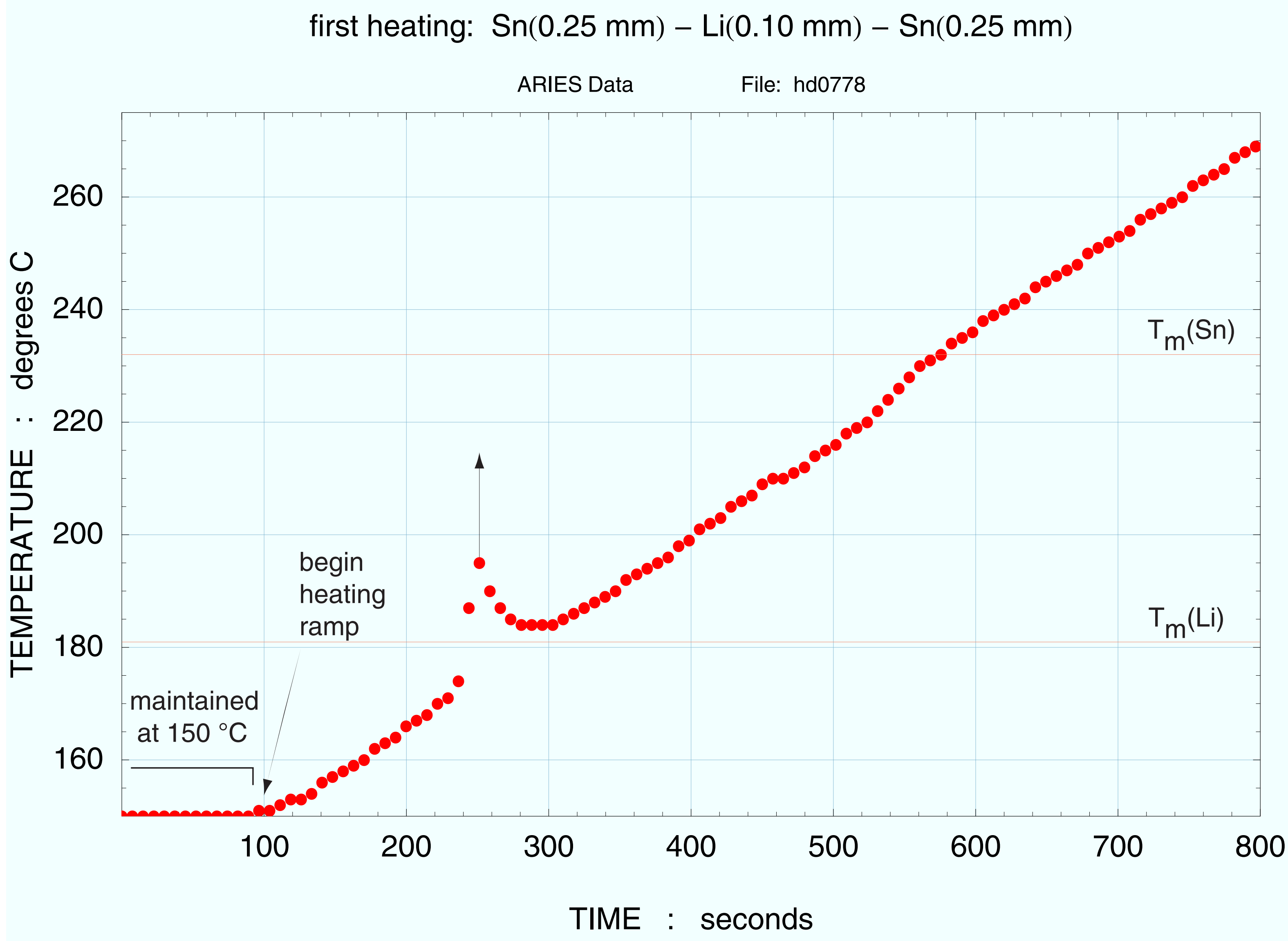
$$m_2 = m_1 \frac{1 + E_S - 2\sqrt{E_S}\cos\theta_S}{1 - E_S - Q_n}$$

(Q_n = inelastic energy loss)

Ion energy spectra identify surface atoms.

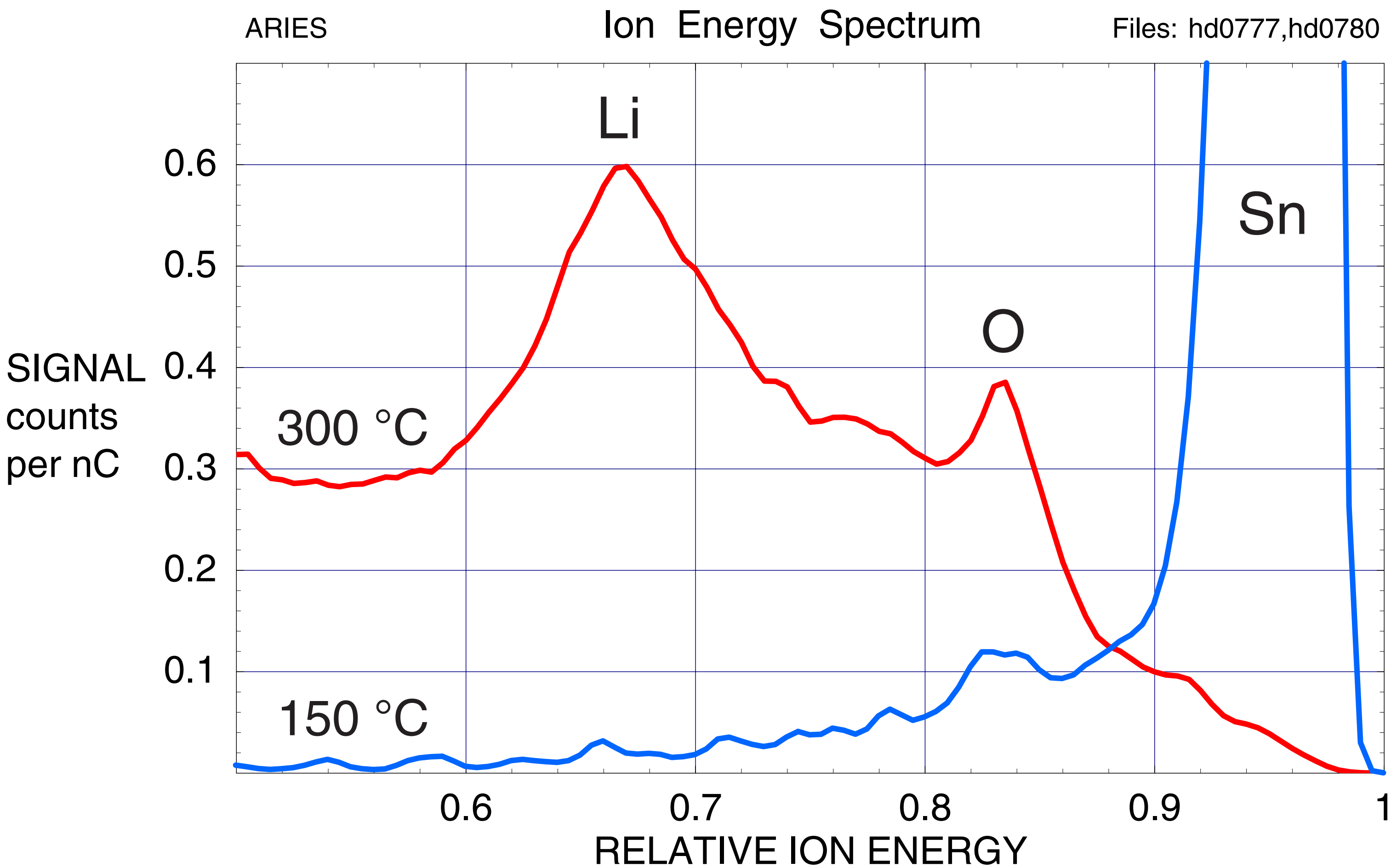


An exothermic reaction occurs near T_m(Li)

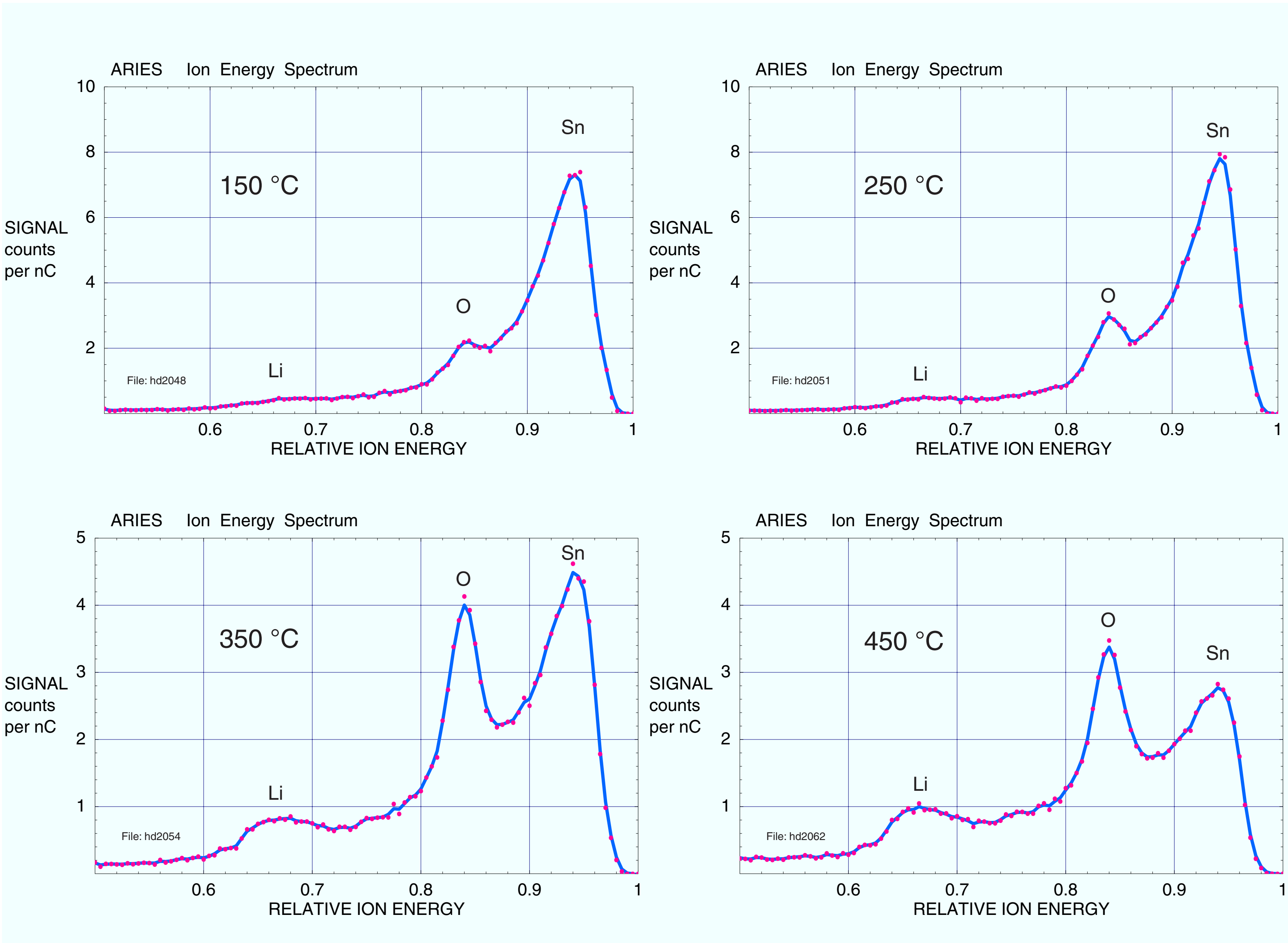


The liquid surface is enriched in Li and O.

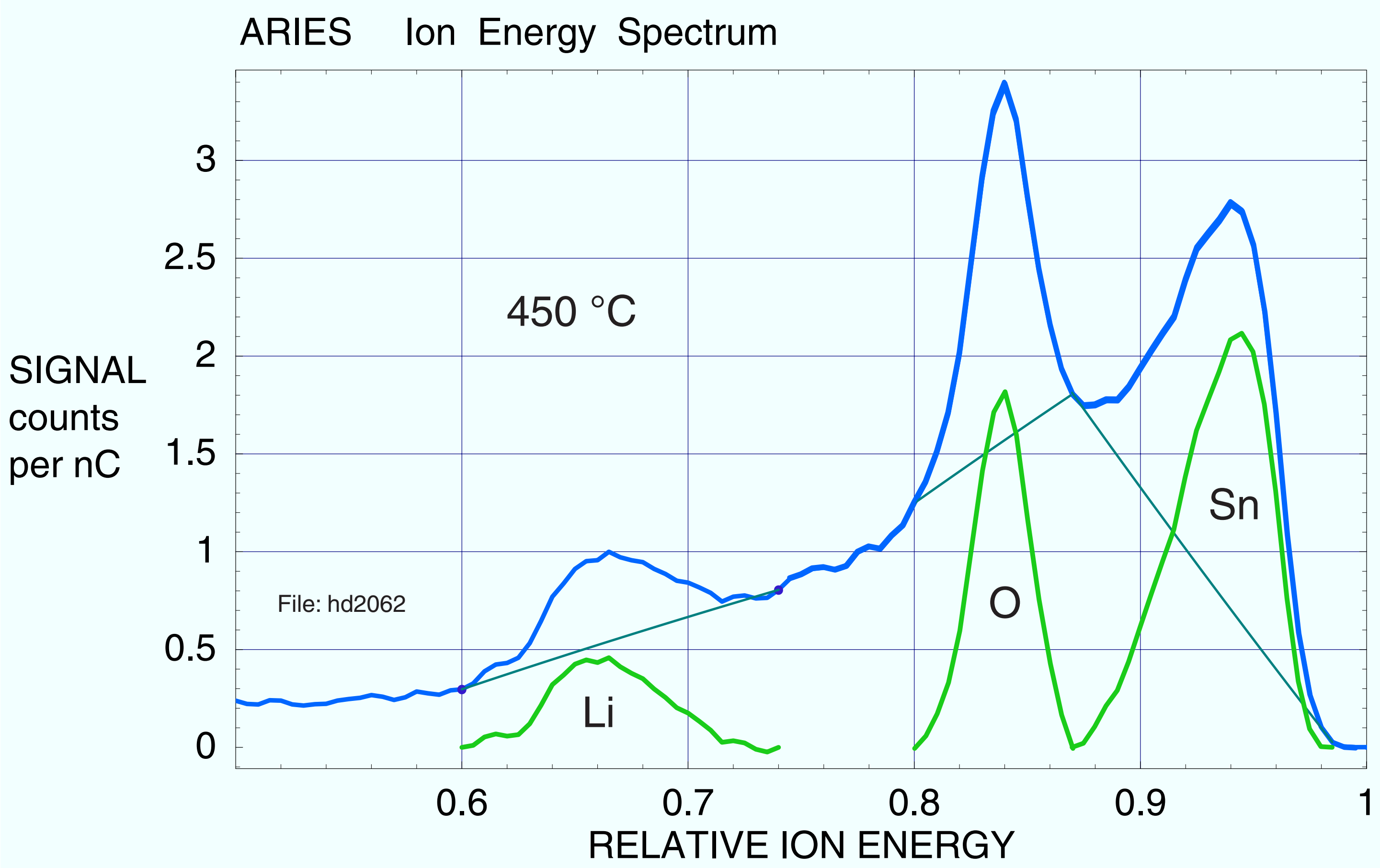
1000 eV He+ --> Sn–Li–Sn
First heating: below and above melting point
Theta = 45.0 deg Alpha = 67.5 deg Temp = 150,300 C



Equilibrium data give Li and O surface coverage.



Peak areas are calculated following subtraction of inelastic background.



Surface concentrations are determined from the scattering cross sections and peak areas.

	Li	O	Sn
• cross sections (10^{-3} Å^2/sr)	9.1	30.9	131.1

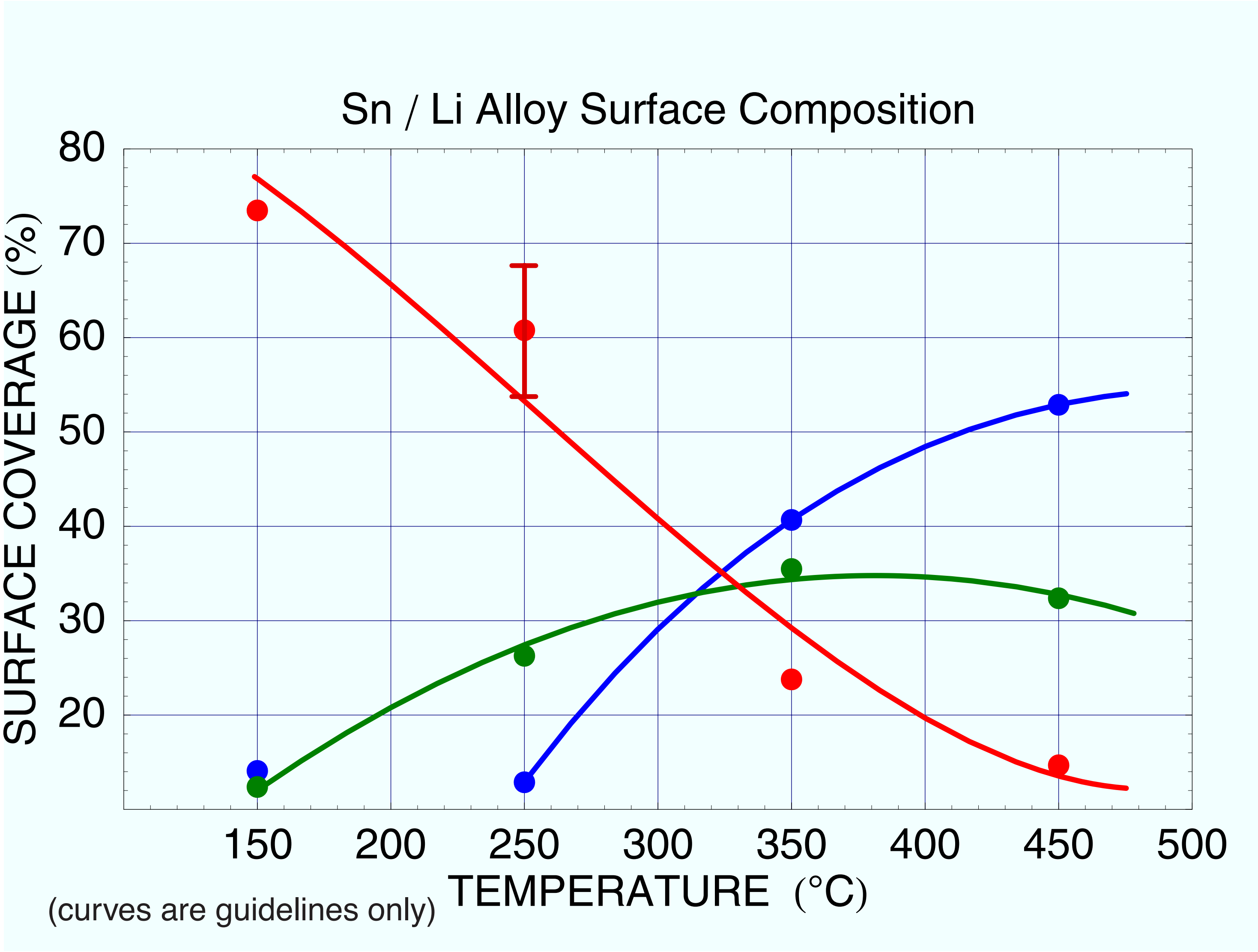
• peak areas

T (°C)	Li	O	Sn
150	0.9	2.7	68.2
250	1.0	7.2	70.5
350	4.5	13.3	37.7
450	5.4	11.2	21.5

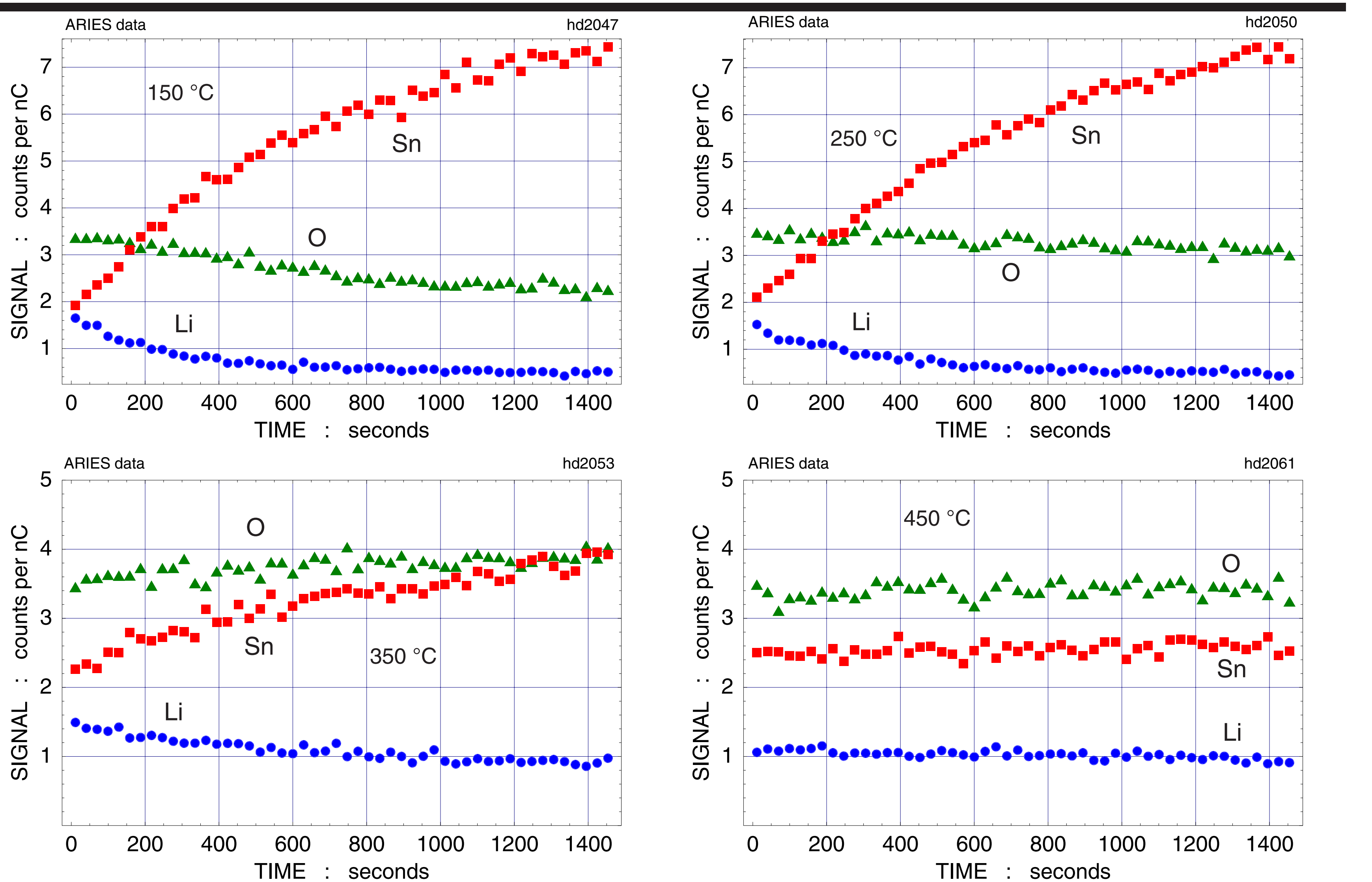
• surface concentrations (atomic %)

T (°C)	Li	O	Sn
150	14.1	12.4	73.5
250	12.9	26.3	60.8
350	40.7	35.5	23.8
450	52.9	32.4	14.7

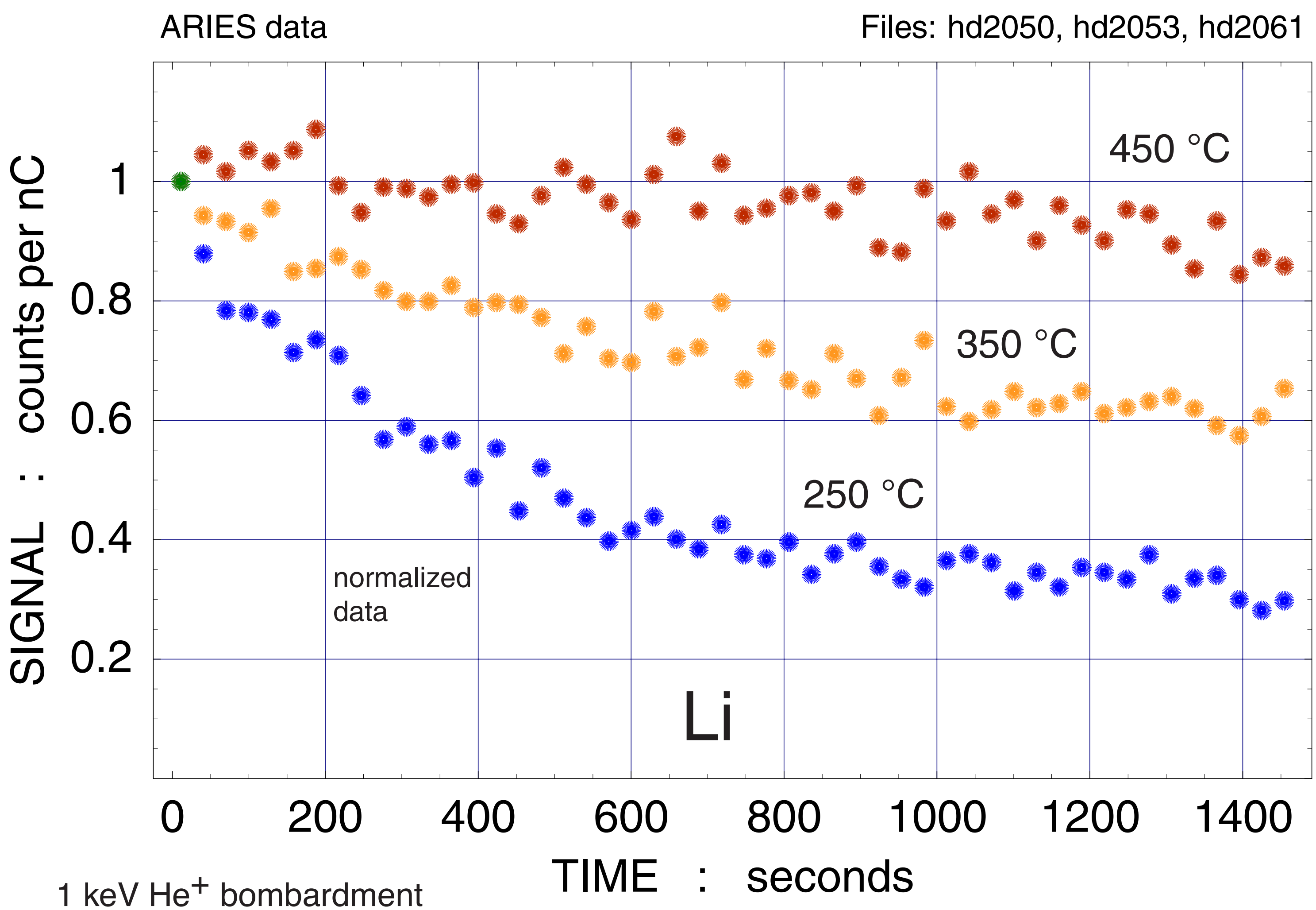
Above 350 °C, the liquid Sn-Li surface is predominantly populated with Li and O atoms.



Transient response depends on temperature.



Surface Li persists longer at higher temperatures.



Model combines effect of segregation, sputtering, and evaporation on surface composition.

- Balance equation:

$$d\theta/dt = J_d - J_s - J_e$$

J_d = Li diffusion flux to surface
J_s = Li sputter flux
J_e = Li evaporation flux.

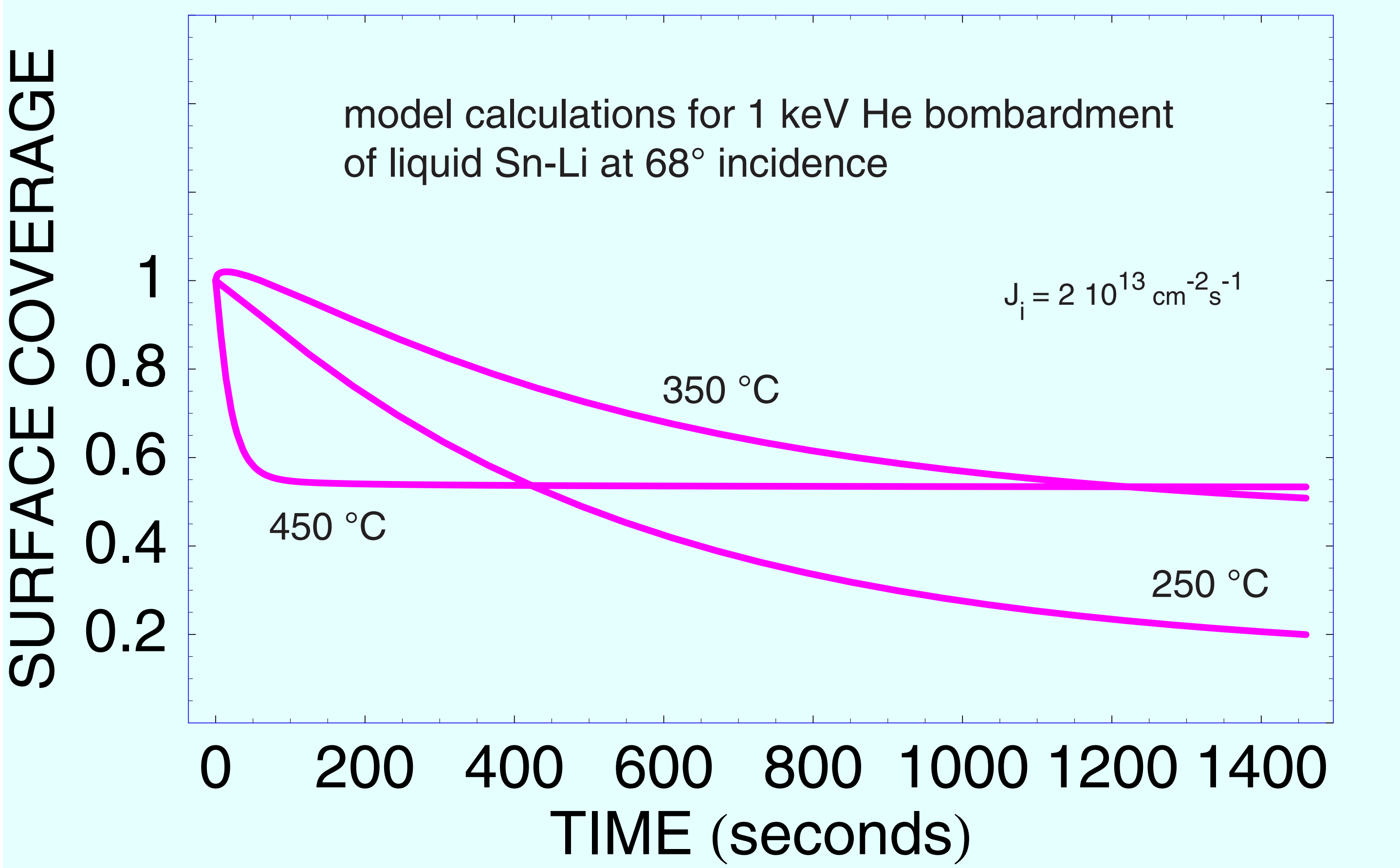
- Time equation:

$$\theta(t) = \frac{e^{-k_1 t}[k_2\sqrt{k_1} + k_3 \mathrm{Erf}(\sqrt{k_1 t})]}{\sqrt{k_1}}$$

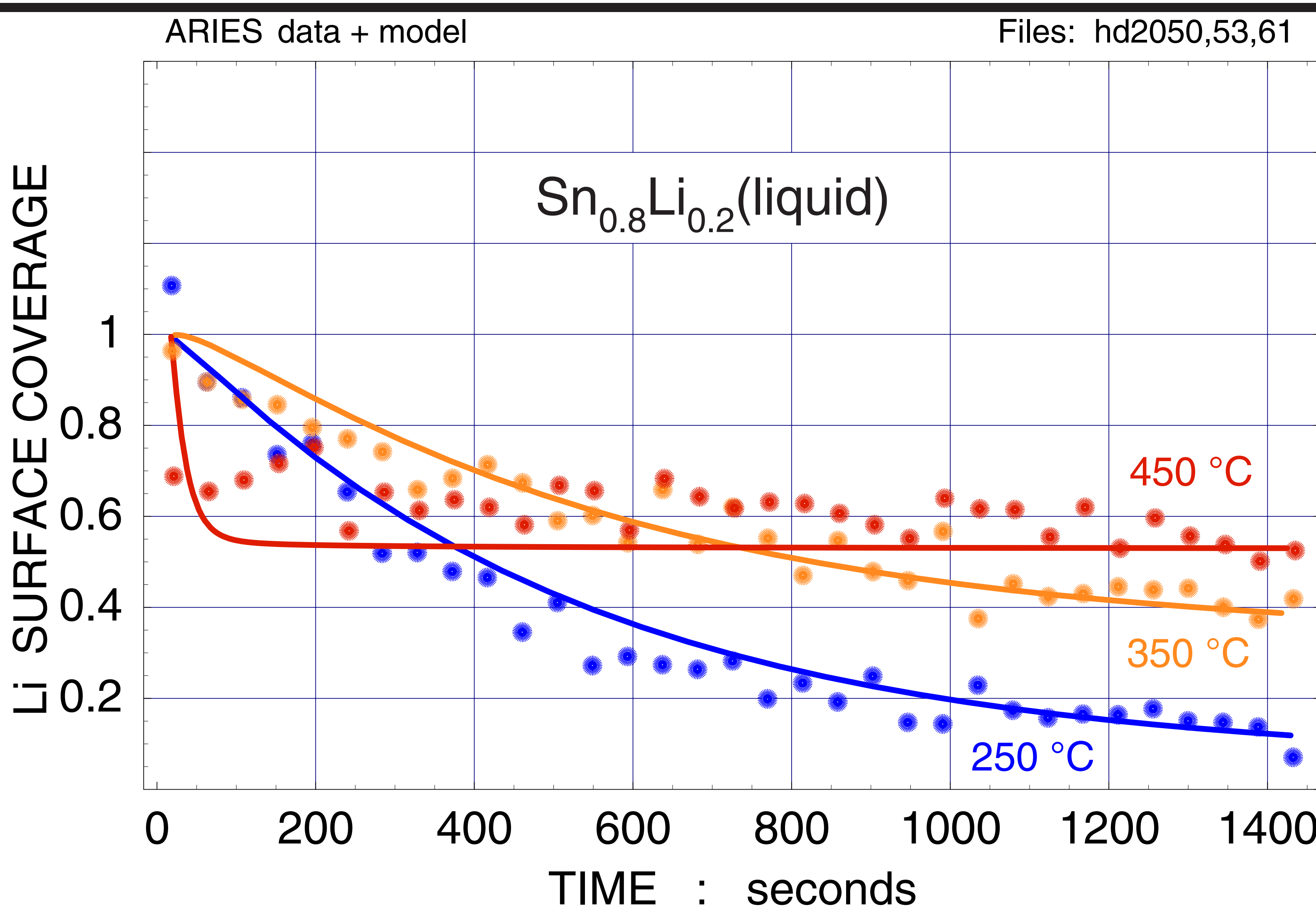
$k_1 = (S_y J_i + J_e)/\theta_0$
$k_2 = \theta(t{=}0)$
$k_3 = c_0\sqrt{D}$
$S_y =$ Li sputter yield

J_i = incident particle flux
θ_0 = surface atom density
c_0 = bulk Li concentration
D = Li diffusivity.

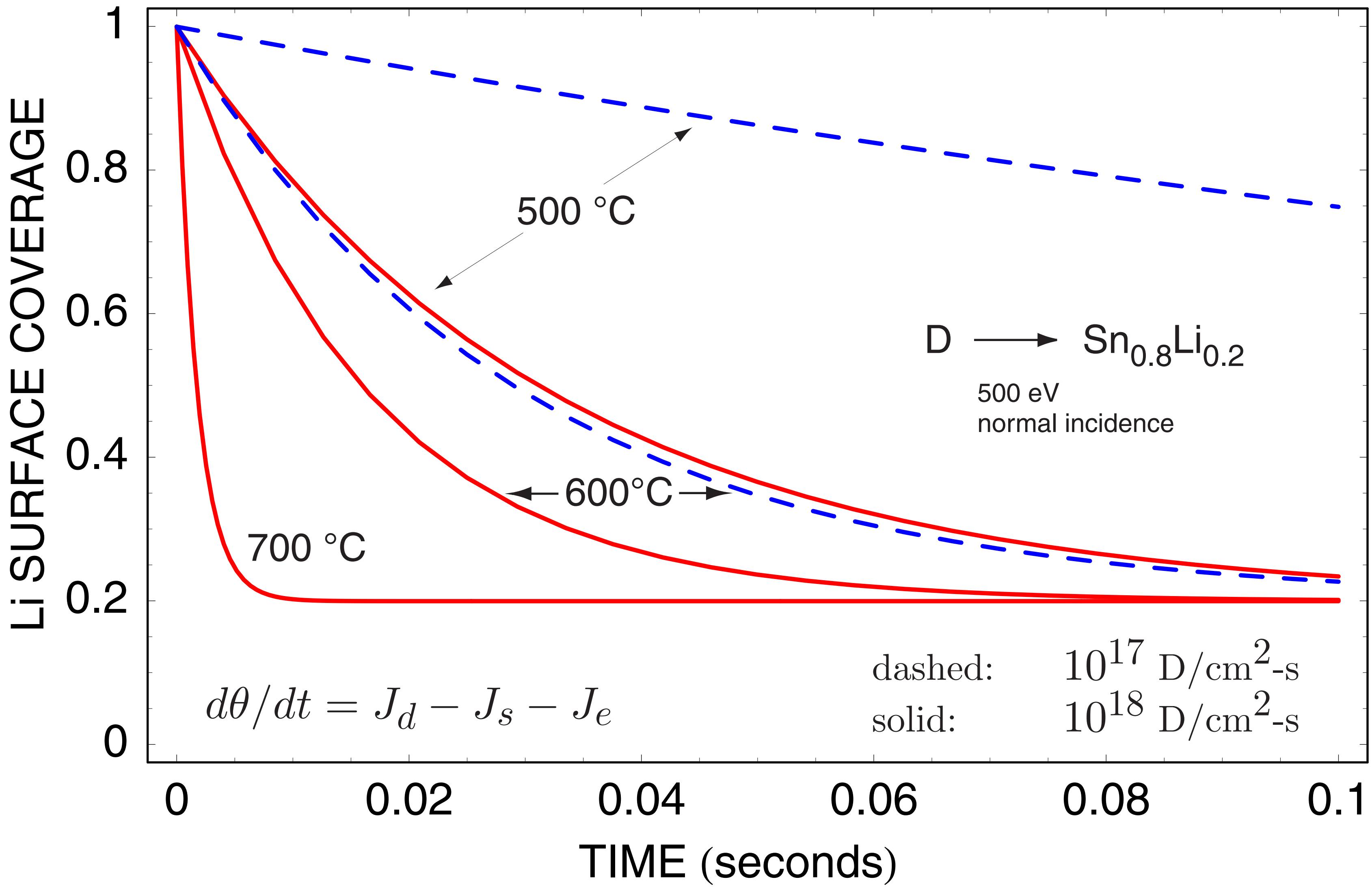
Modeling time dependence of Li surface coverage.



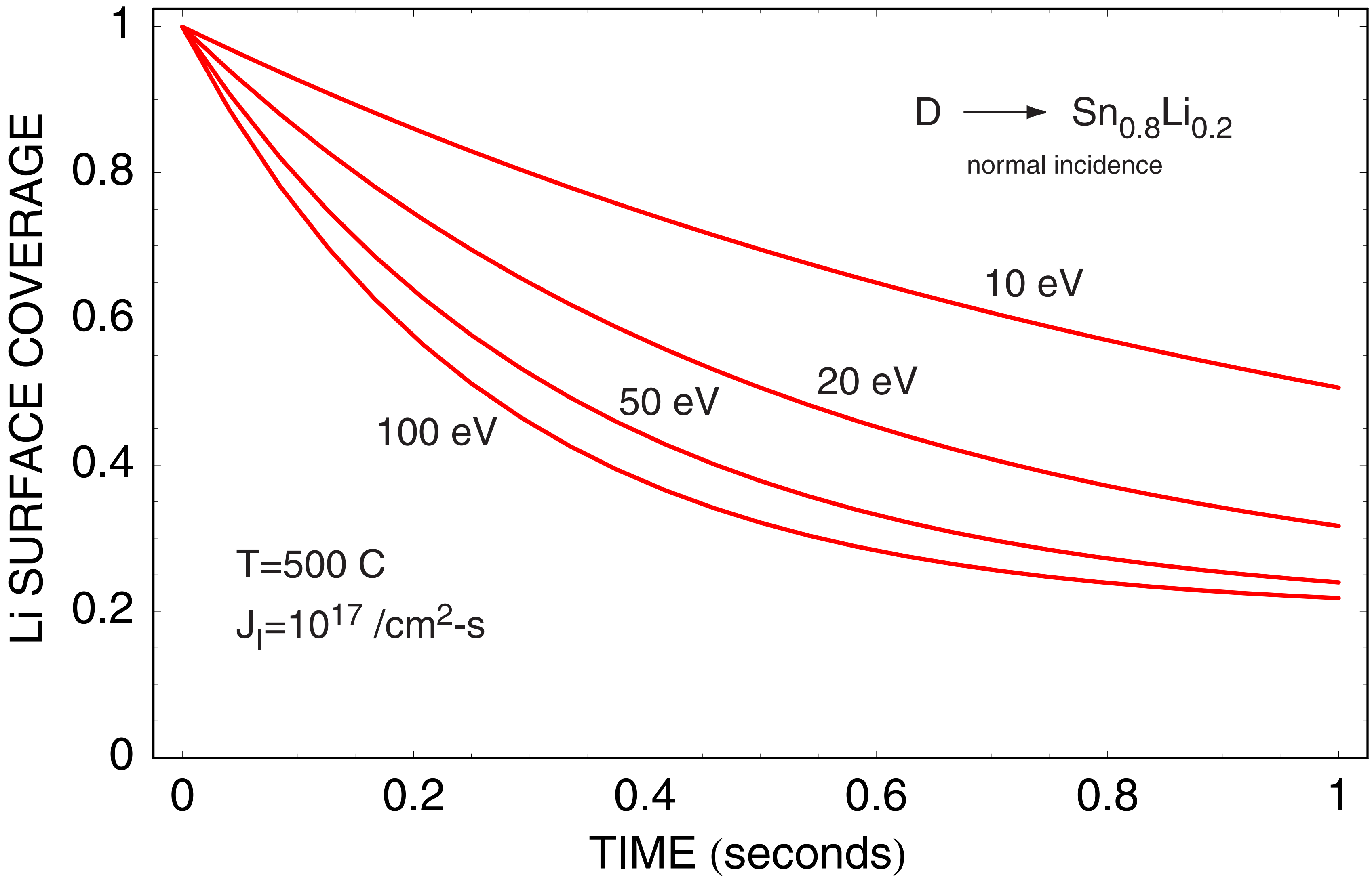
Model and data agree except near t=0 at 450 °C.



At high temperature or incident particle flux the surface Li layer does not persist.



At moderate temperature, surface Li concentration strongly depends on incident particle energy.



Summary

1. Surface composition measurements show that Li and O segregate to the free surface of liquid Sn-Li.
2. At 450 °C, Li evaporation appears to nearly equal the segregation rate.
3. Modeling the surface composition indicates that the lithium surface layer will survive for short times in a fusion plasma, depending strongly on incident flux, particle energy, and surface temperature.

$\Rightarrow$ Using a Sn-Li alloy or adding Li to pure Sn may be of benefit in enriching the plasma-facing surface with Li, if the surface exposure time is kept sufficiently short.

Planned work

- Laboratory studies will focus on liquid Li:
  - transient and equilibrium measurements of surface composition will be used to evaluate
    - oxygen segregation and sputtering,
    - deuterium adsorption and release.
- Simulation studies will model the liquid surface:
  - the multilayer TRVMC98 code will be used to calculate sputter yields of liquid surfaces of appropriate composition and density.