Diffusion in Al-Ni-Ce Melts

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Motivation

- Influence of structure and thermodynamic forces on diffusion in metallic melts?

$$D_{ik} = (D^*_i N_k + D^*_k N_i)\Phi M \quad \textit{in melts?}$$

L.S. Darken, Trans. AIME 180 (1948) 430

J.R. Manning, Phys. Rev. 124, 2 (1961) 470

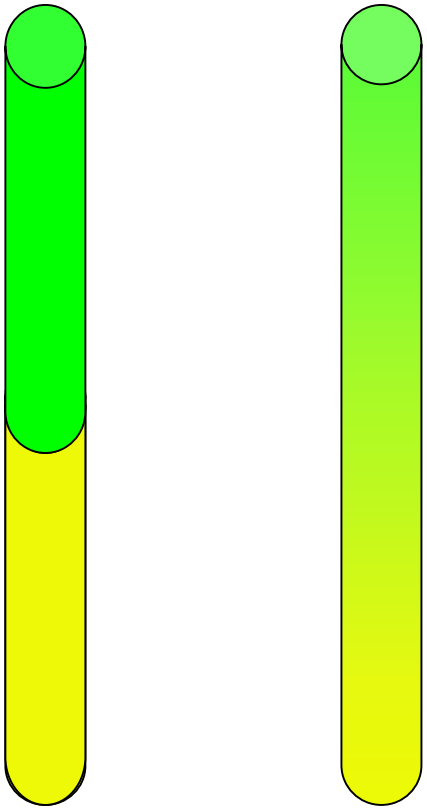
ambiguous result in SnIn melts: $M \neq 1$

G. Frohberg in *Scientific Results of the German Spacelab Mission D-2*, DLR Cologne (1994) 275ff

- $AlNi_{20}$, $AlNi_{20}Ce_3$, $AlNi_{10}Ce_3$

Long-Capillary Technique

∅1,5 mm x 30 mm

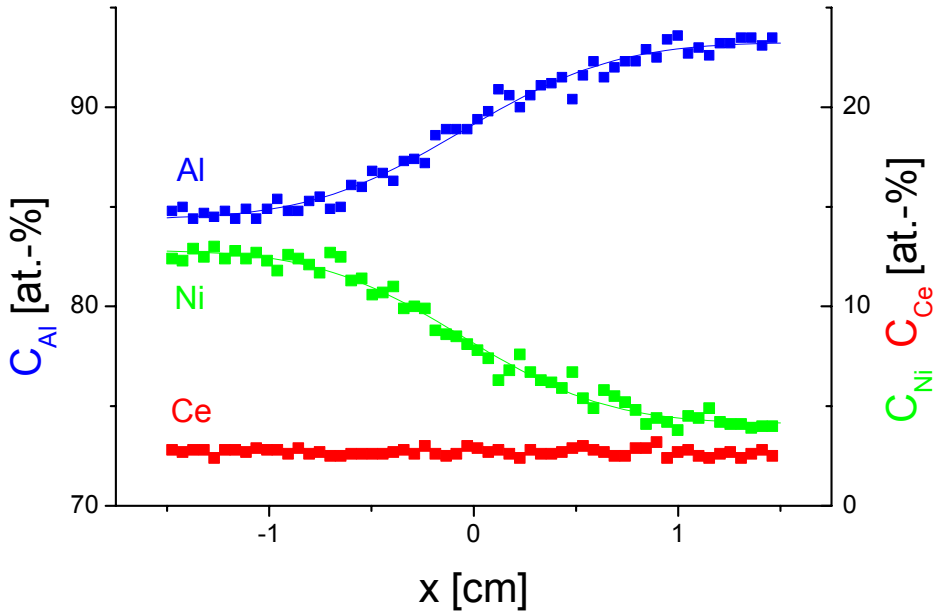


t = 0
T = RT
ΔC = 10 at%

t > 0
T > T_{liq}

EDS
AAS

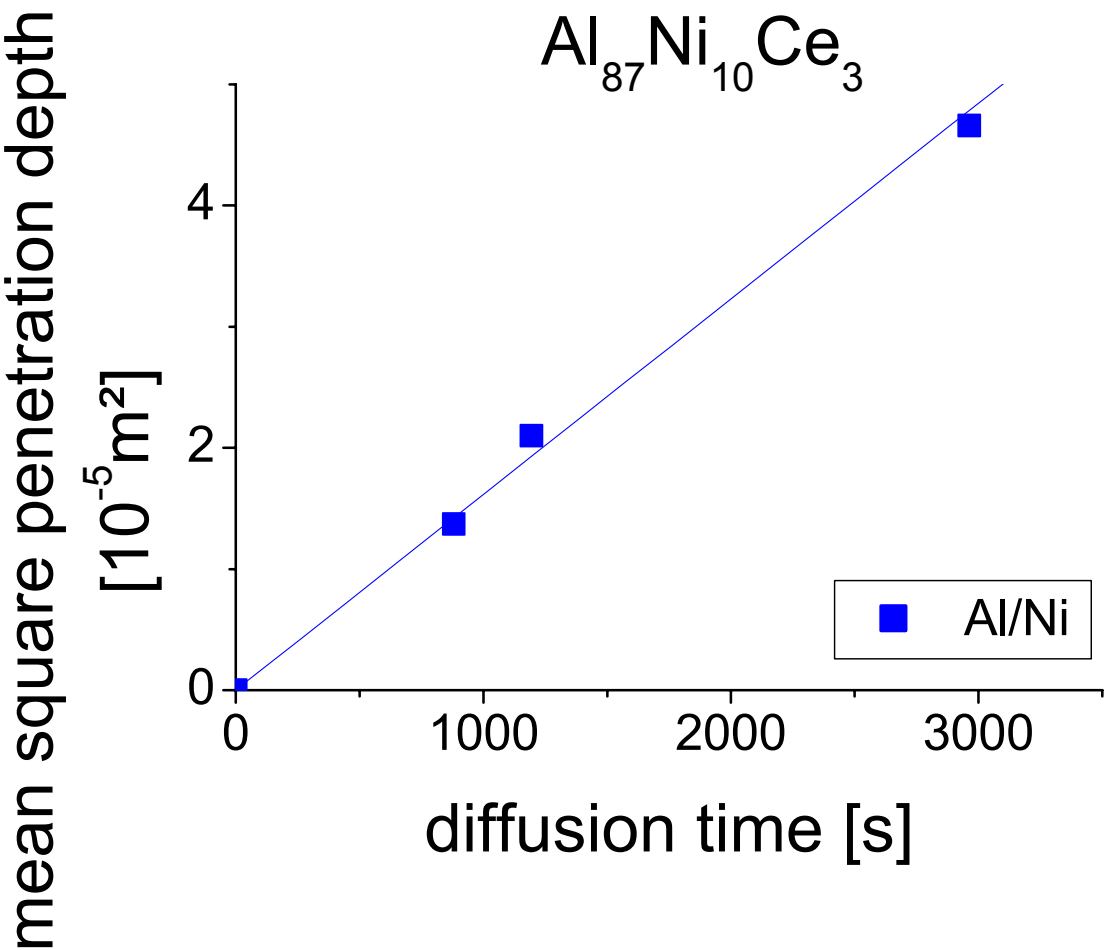
ICP-MS



Corrections:
- sample length
- effective diffusion time

D_{ik} = fit parameter
relative error = ±30...40%

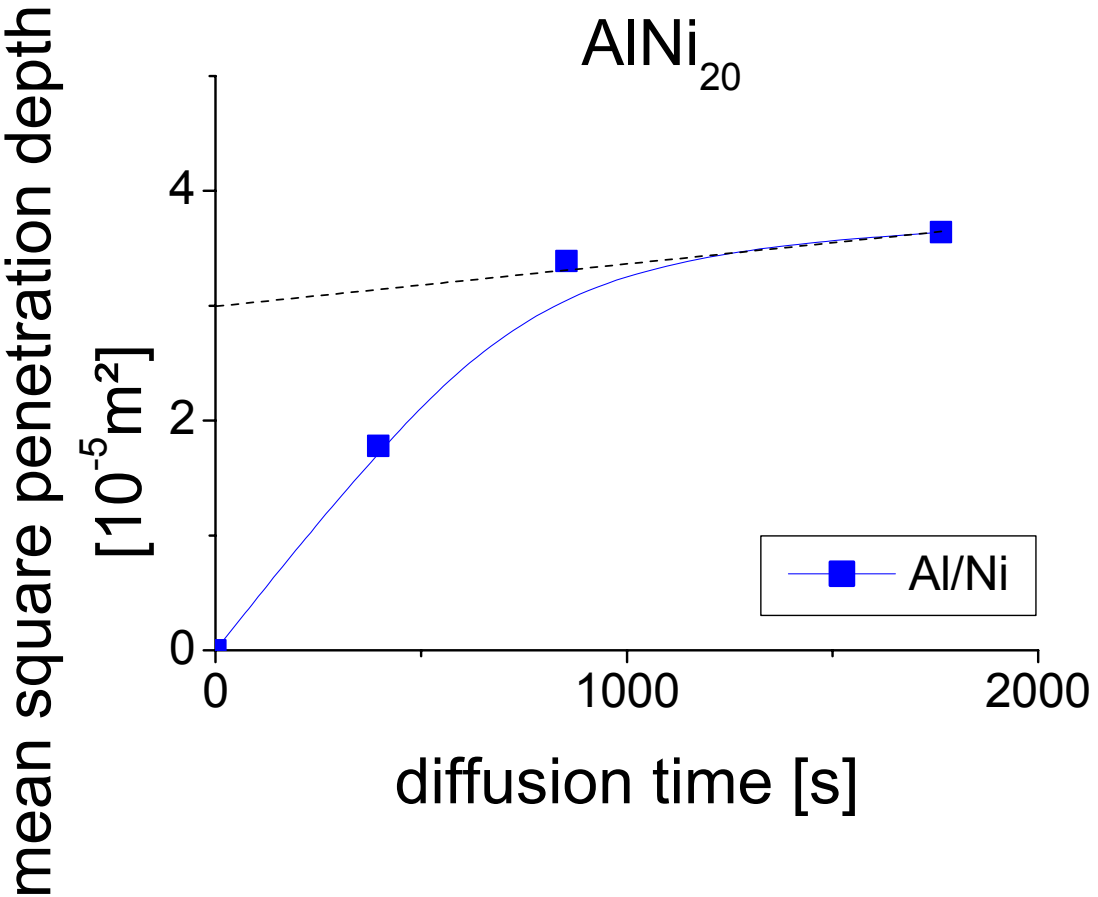
Check for additional Transport



- $x^2 = 2Dt$

$\Rightarrow$ **no additional transport**

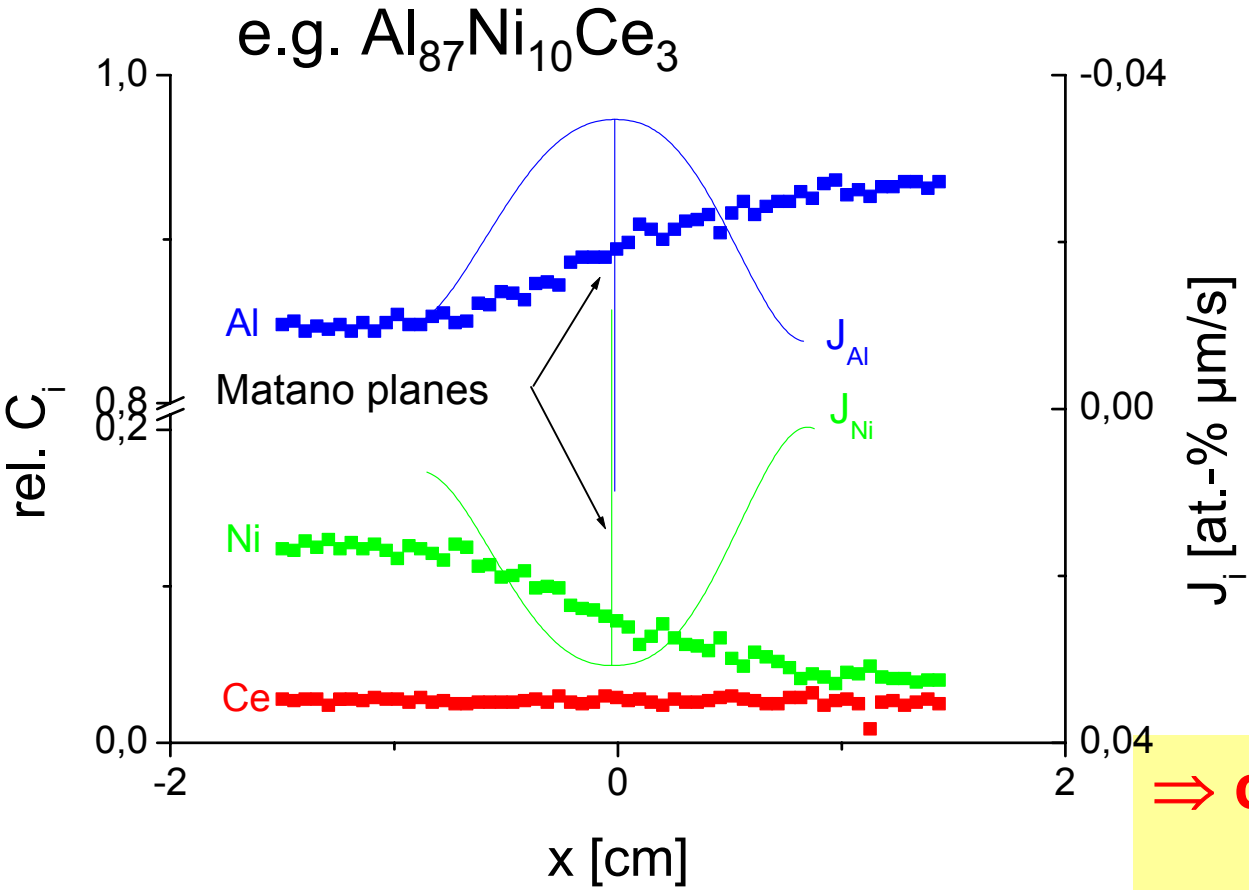
Check for additional Transport



- $x^2 = 2Dt + x^2_0$

$\Rightarrow$ **additional transport segregation Al_3Ni_2**

Boltzmann-Matano Analysis



MultiDiFlux

M.A. Dayananda, Purdue Uni.

⇒ conc. independence of *D*

- **Matano planes at x = 0**
- **$J_{Al} + J_{Ni} = 1$**

⇒ quasi-binary

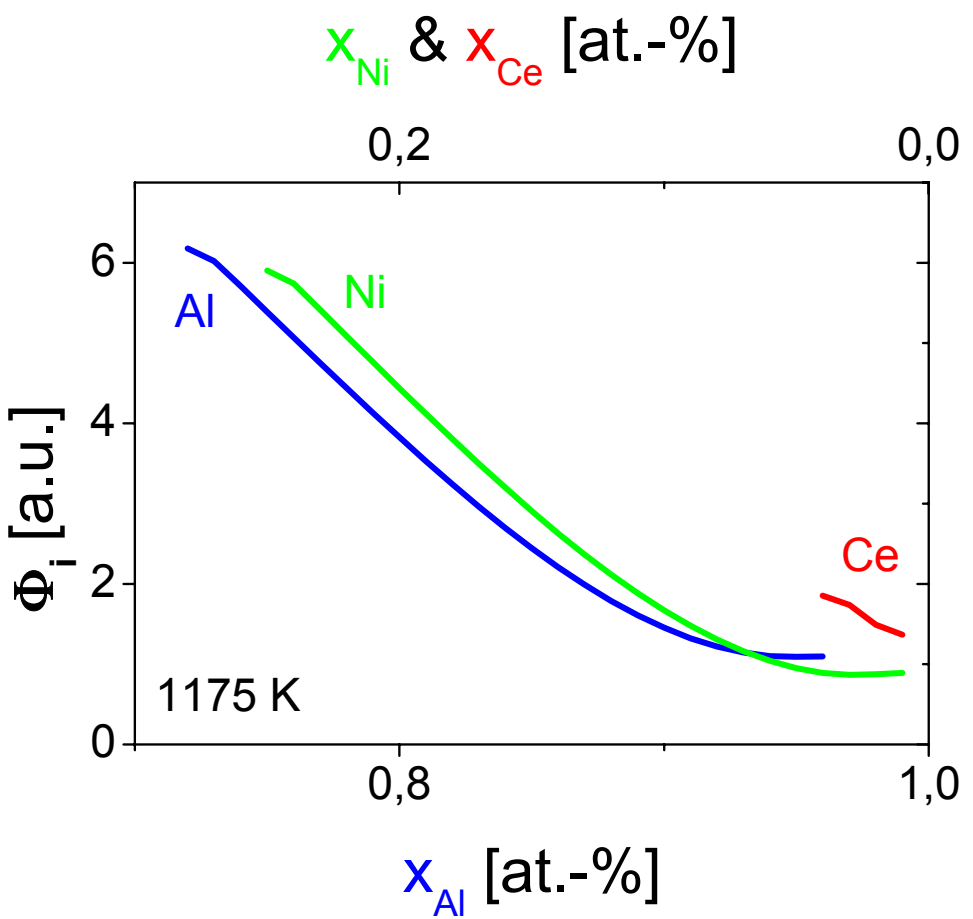
Thermodynamic Factor Φ

Pandat

R. Schmid-Fetzer / Uni. Clausthal

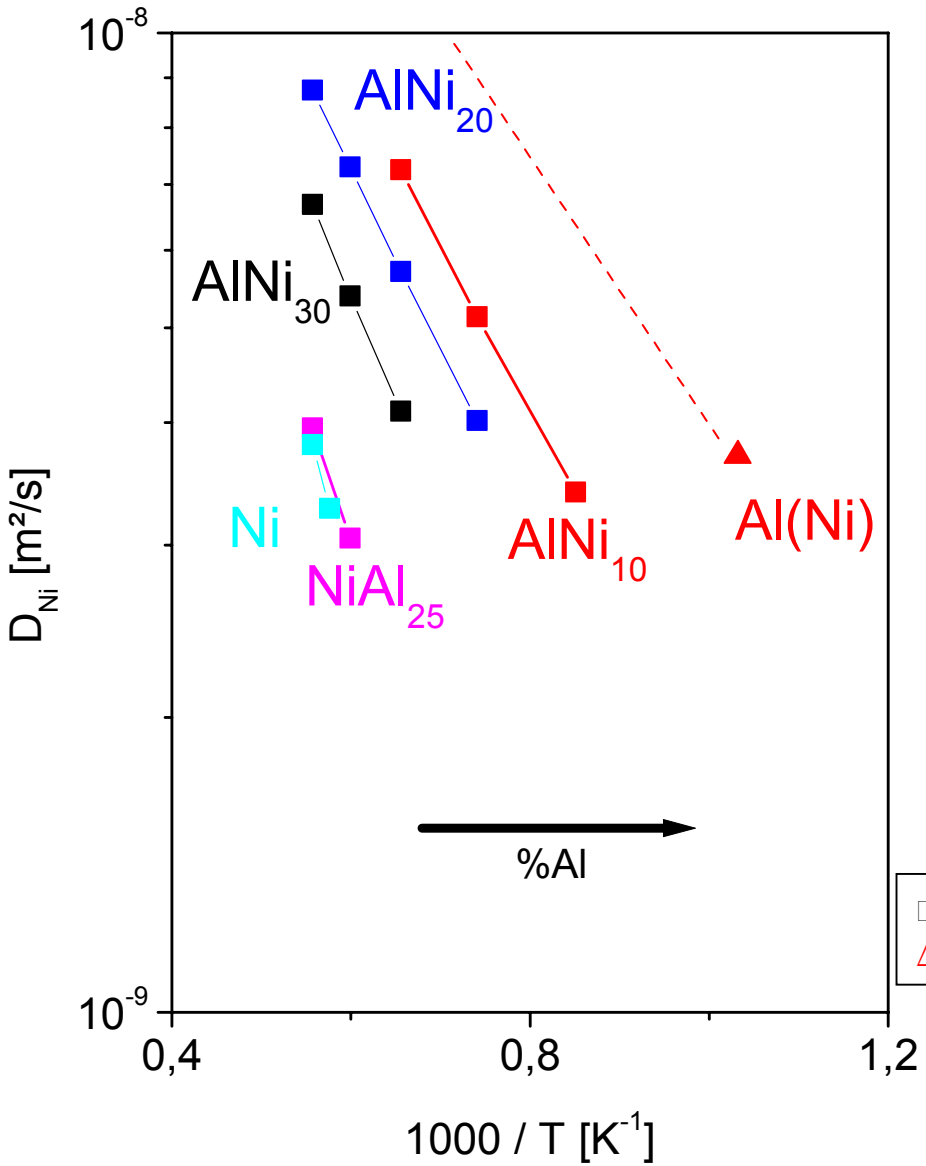
$$\mu_i = \mu_{0,i} + RT\ ln(a_i)$$

$$\Phi_i = 1 + (d\ ln(\gamma_i) / d\ ln(x_i))$$



- **Φ_i depends strongly on alloy composition**

Ni Self Diffusion in AlNi

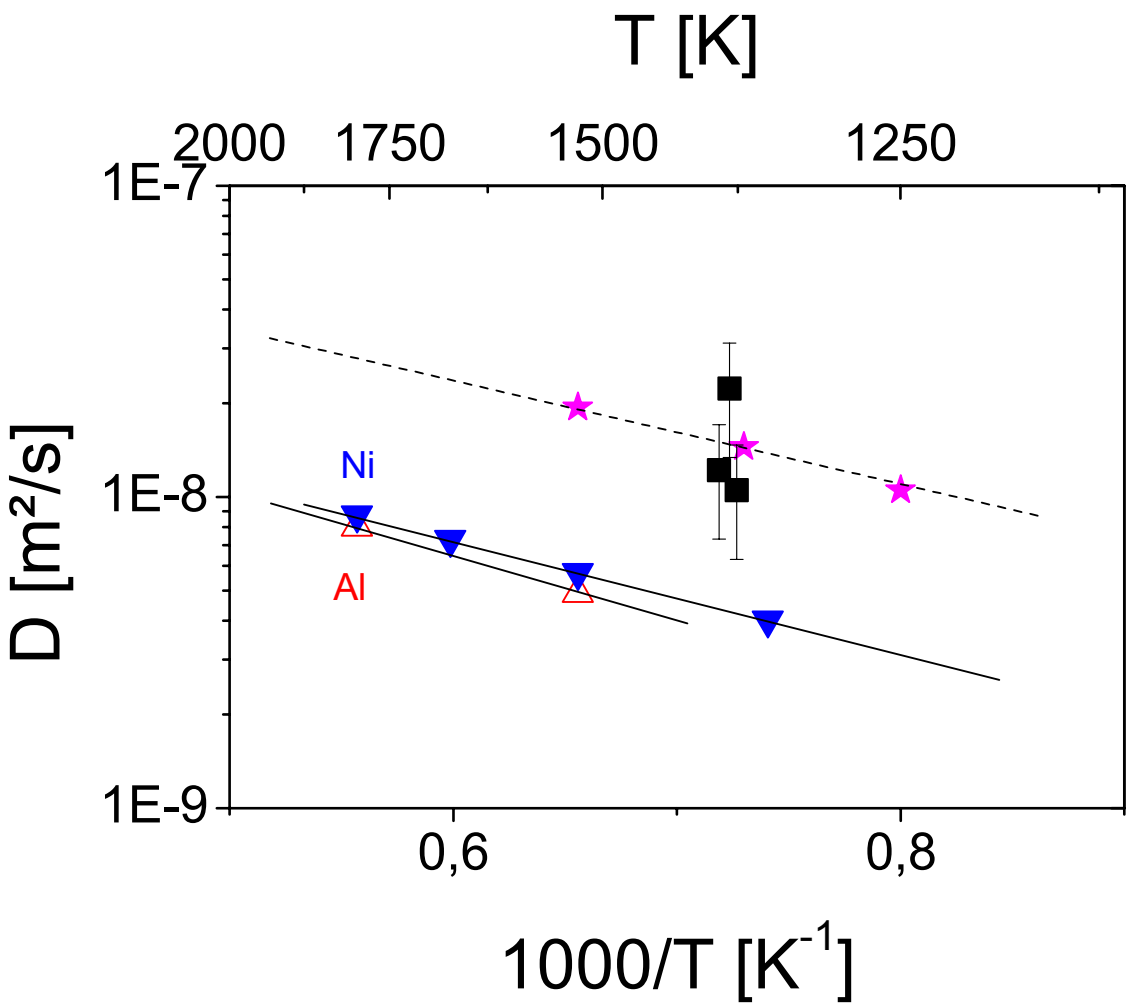


- **increase of D^*_{Ni} with x_{Al}**

⇒ influence of CSRO

□ Horbach, Meyer et al., APL 82,1 (2005) 11918
Δ Garandet, Griesche et al, I. J. Thermophys. 25, 1 (2004) 249

Darken-Test in $AlNi_{20}$

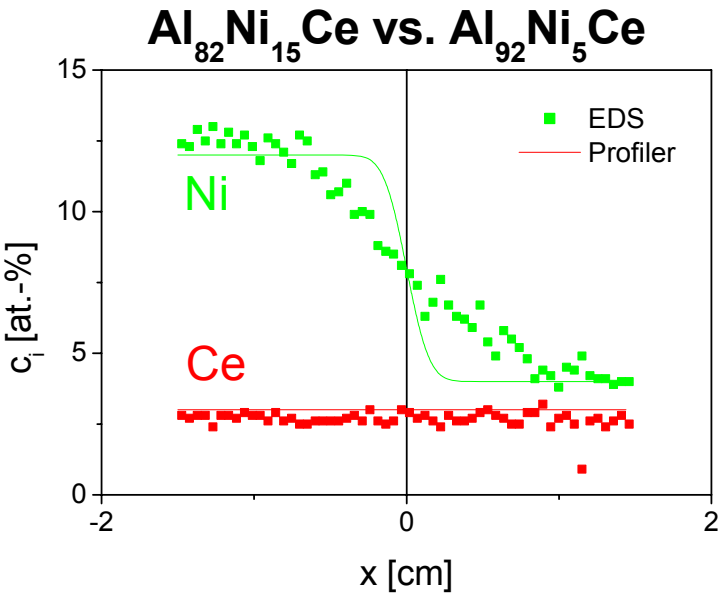


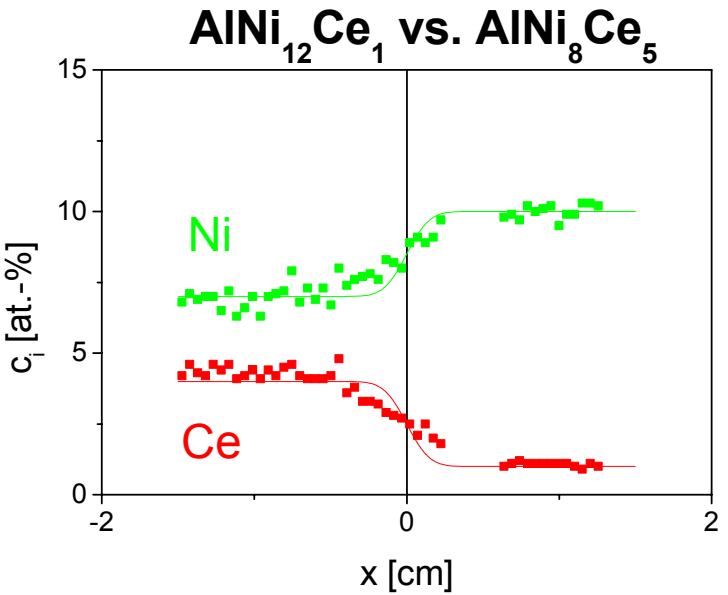
- ***calc.*** D_{AlNi} **= exp.** D_{AlNi}

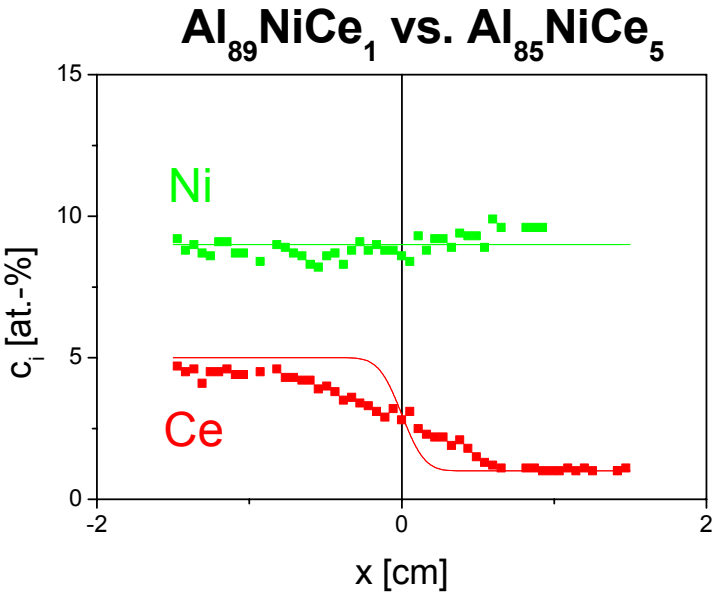
⇒ **Darken fulfilled**

▽ △ Horbach, Meyer et al., APL 82,1 (2005) 11918
■ exp. interdiffusion, this work
★ calc. w/ Darken-Eq., M=1

Darken-Test in $Al_{87}Ni_{10}Ce_3$







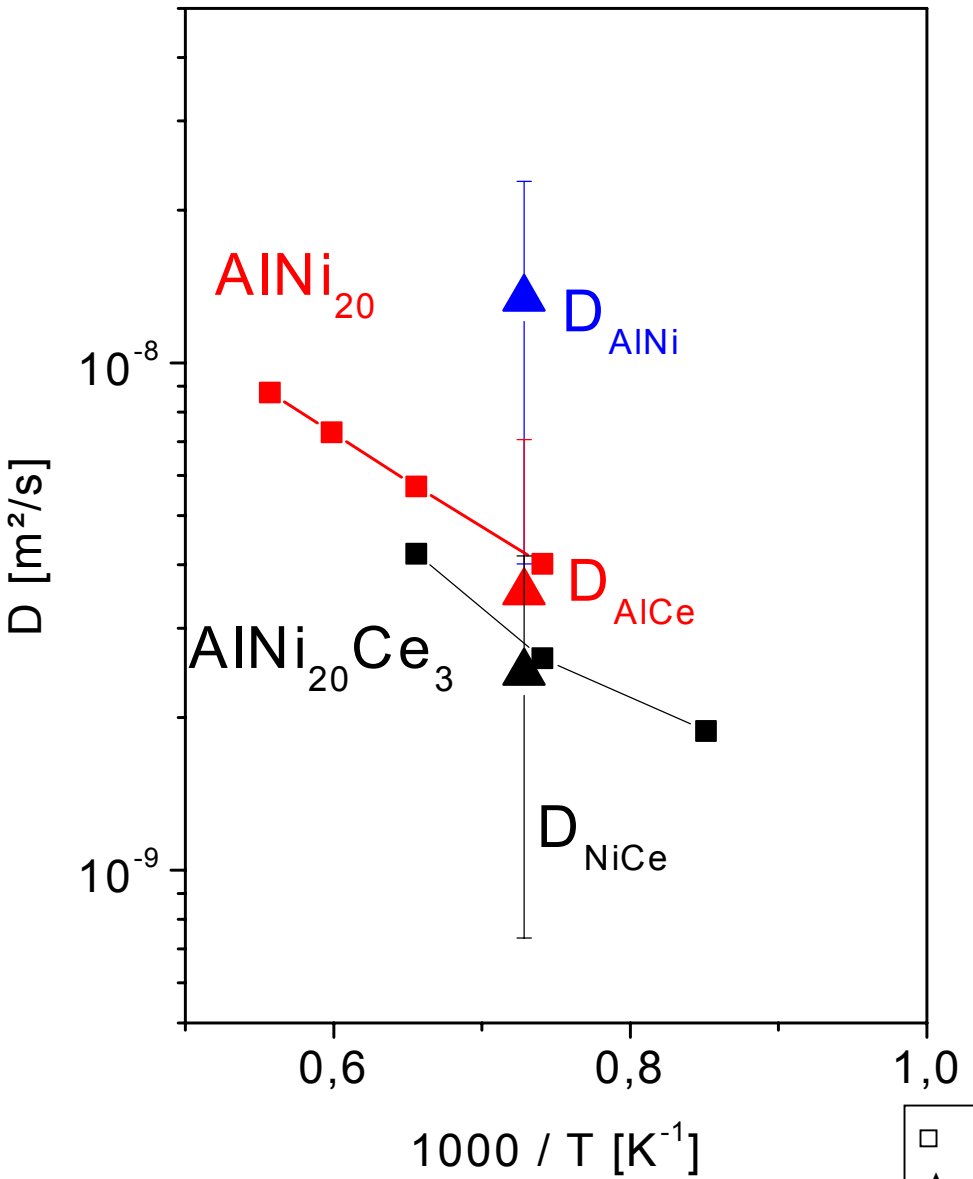
PROFILER

J. Morral / Uni. Connecticut

- **no singularity**
- **poor agreement**

⇒ Darken fulfilled?

Diffusion in $Al_{77}Ni_{20}Ce_3$



- **Ce decreases D^*_{Ni} (~30%)**

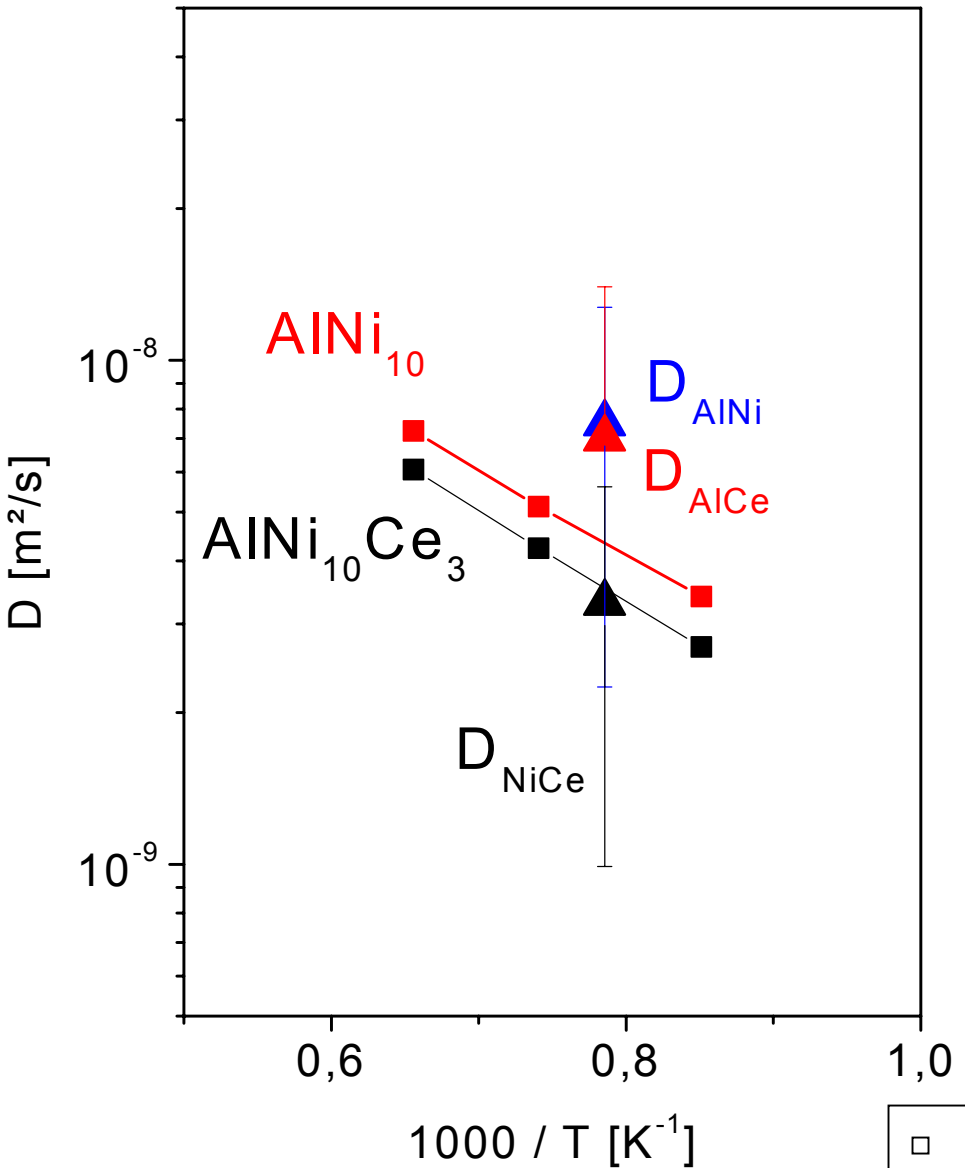
⇒ structural change

- **$D_{AlNi} > D_{AlCe} > D_{NiCe}$**

- **$D_{AlNi} \approx 5\ D^*$**

⇒ thermodynamic forces

Diffusion in $Al_{87}Ni_{10}Ce_3$



- **Ce decreases D^*_{Ni} (~20%)**

⇒ structural change

- **$D_{AlNi} > D_{AlCe} > D_{NiCe}$**
- **$D_{AlNi} \approx 2\ D^*$**

⇒ thermodynamic forces

Relationship Diffusion - Thermodynamic Force

	ratio D_{ik} / D*	***Φ***
$AlNi_{20}$	$D_{AlNi} \approx 3{,}8\ D^*$	3,8
$AlNi_{20}Ce_3$	$D_{AlNi} \approx 5\ D^*$	4
$AlNi_{10}Ce_3$	$D_{AlNi} \approx 2\ D^*$	1,8

- $D_{ik} \approx D^* \Phi$

J. Bøttiger et al., Mat. Sci. Eng. A 178 (1994) 65

Conclusion

- ✓ Ce addition decreases $D \Rightarrow$ change in structure
- ✓ D^* depends strongly on AlNi composition $\Rightarrow$ CSRO
- ✓ Φ depends strongly on alloy comp. $\Rightarrow$ strong atomic interaction
- ✓ validation of Darken‘s equation in molten $AlNi_{20}$
- ✓ qualitative validation of Darken‘s equation in molten AlNiCe?

  (check $Al_{77}Ni_{20}Ce_3$)
- ✓ rule-of-thumb: ratio of D_{ik} / D^* correlates with Φ

End

Acknowledgement
- DFG SPP 1120
- Auswärtiges Amt