Density and viscosity measurement of liquid alloys

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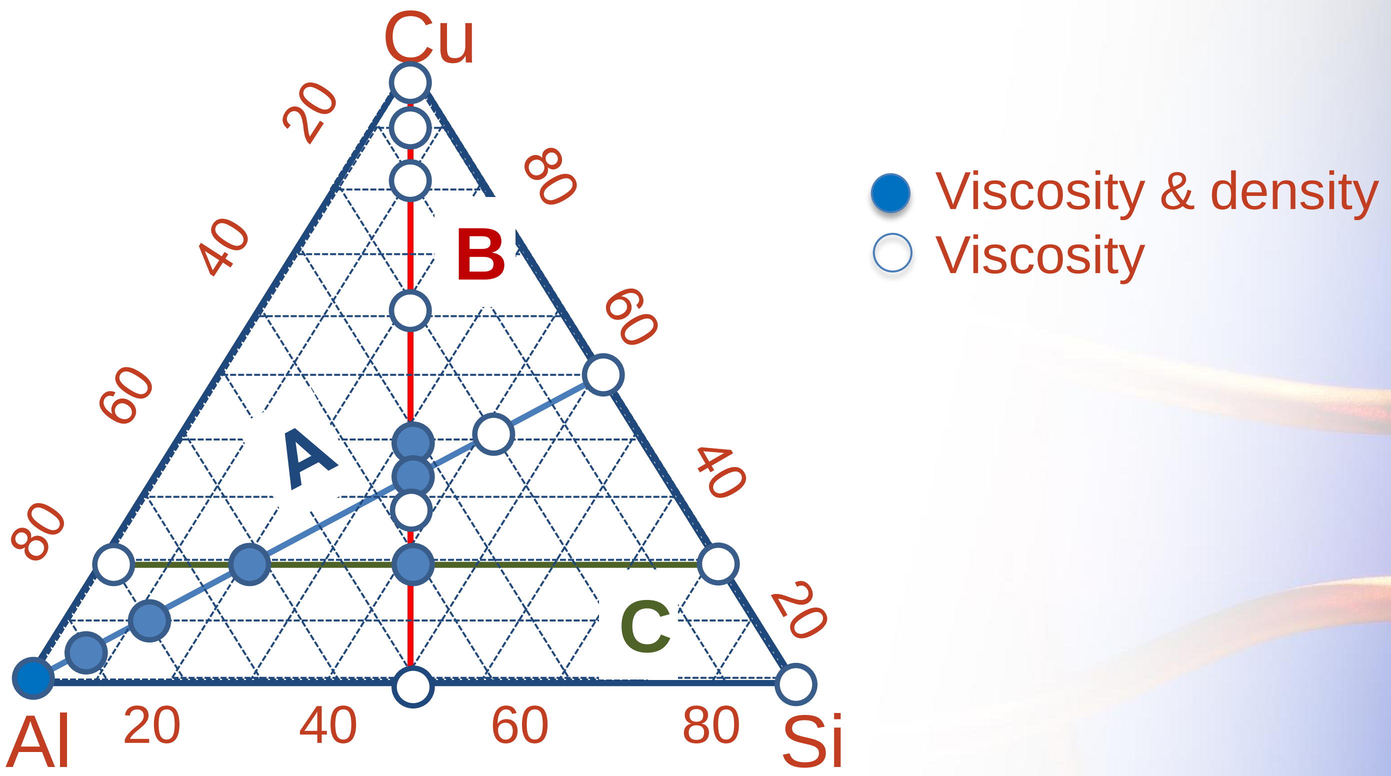
Goal

Establishment of the link between thermodynamic and thermophysical properties of the liquid phase density, viscosity and surface tension

Thermodynamic properties	Thermophysical properties
Molar volume: *V*	Density
Excess Gibbs energy: ^{E}G	Viscosity
	Surface tension

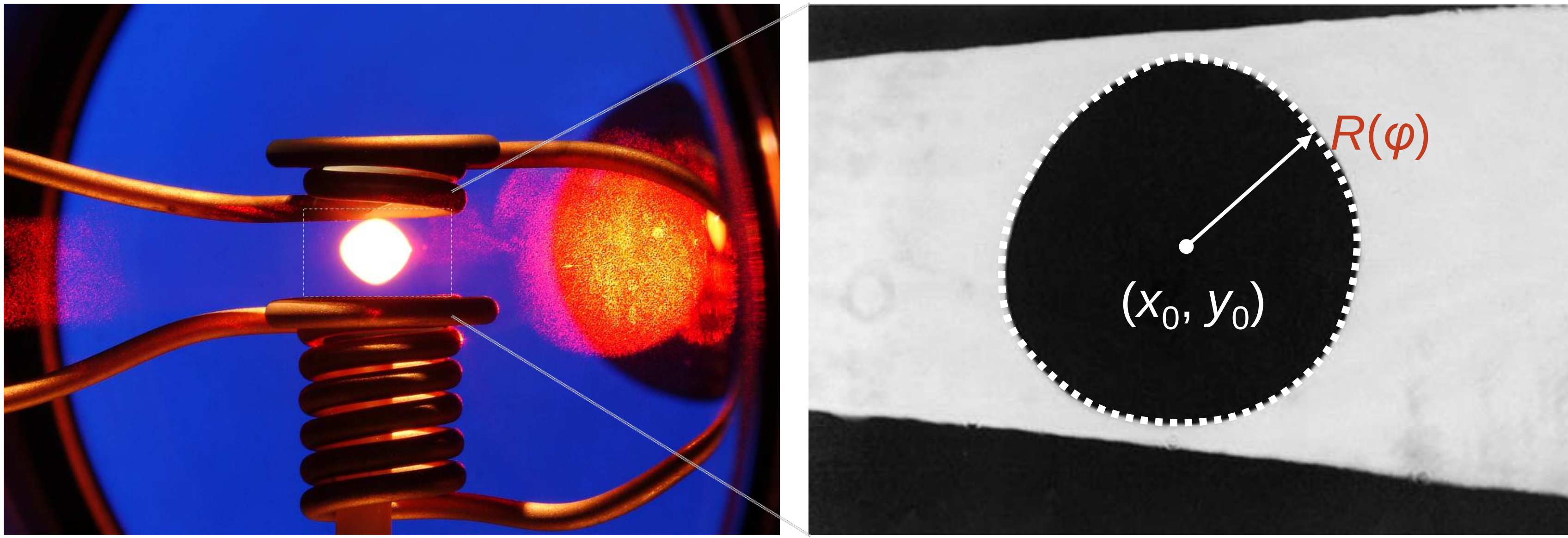
Al-Cu-Si

investigated compositions for density and viscosity



measurements carried out in the Al-rich corner

Density measurement



Edge fit using Legendre polynomial

$$\langle R(\varphi)\rangle = \sum_{i=0}^{6} a_i P_i(\cos(\varphi))$$

Volume calculation

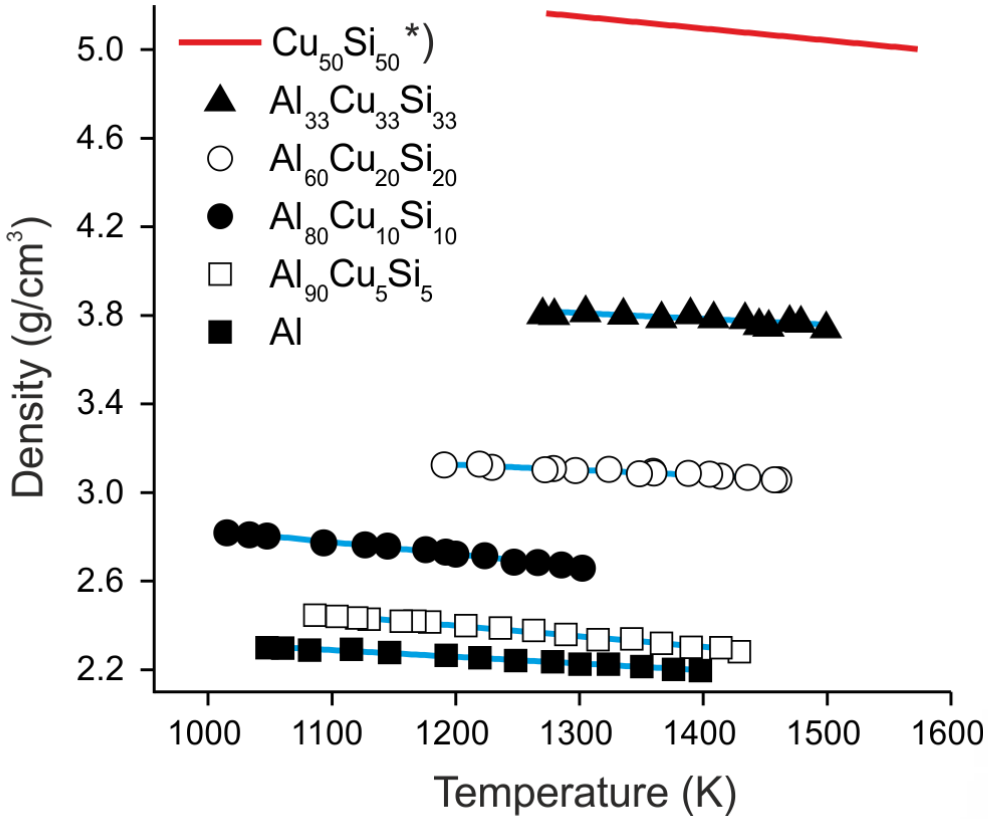
$$V_P = \tfrac{2}{3}\pi \int_0^{\pi} < R(\varphi) >^3 \sin(\varphi)\, d\varphi$$

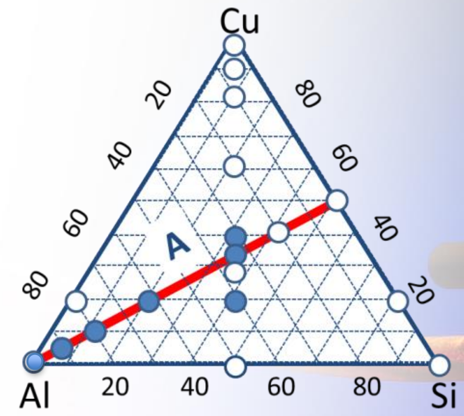
$(\ \Delta\rho/\rho \approx +/- 1\%\ )$

Containerless processing can achieve

- high temperatures and
- investigation of highly reactive materials

Al-Cu-Si - density





ρ is linear in temperature over $T_L < T < T_L + 500$ K

*) Adachi, Schick, Brillo, Egry Watanabe, J. Mater. Sci. 45 (2010)

Molar volume ***(V = M / ρ)*** of a mixture

Ideal volume

$$V_{ideal} = \sum c_i V_i$$

Real volume

$$V_{real} = V_{ideal} + \Delta V$$

Simple expression

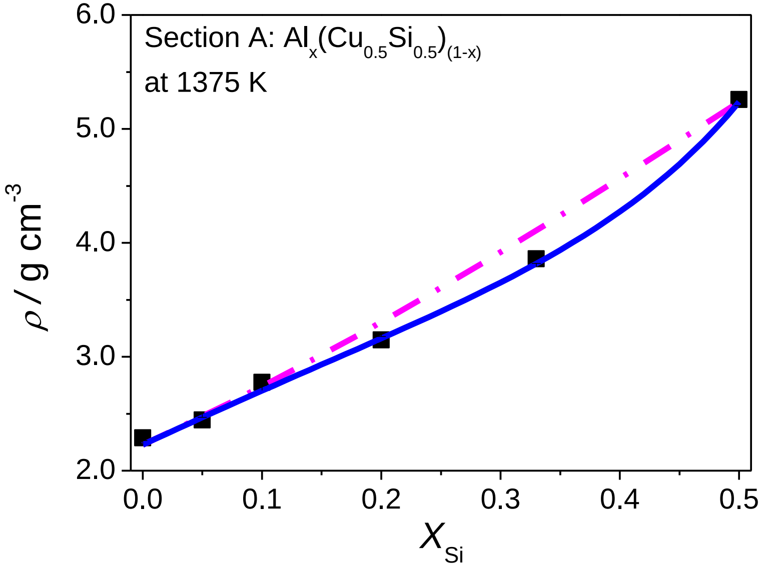
$$\Delta V = \sum_{i}^{2} \sum_{i<j}^{3} c_i c_j V^{i,j} + c_1 c_2 c_3 {}^{T}V$$

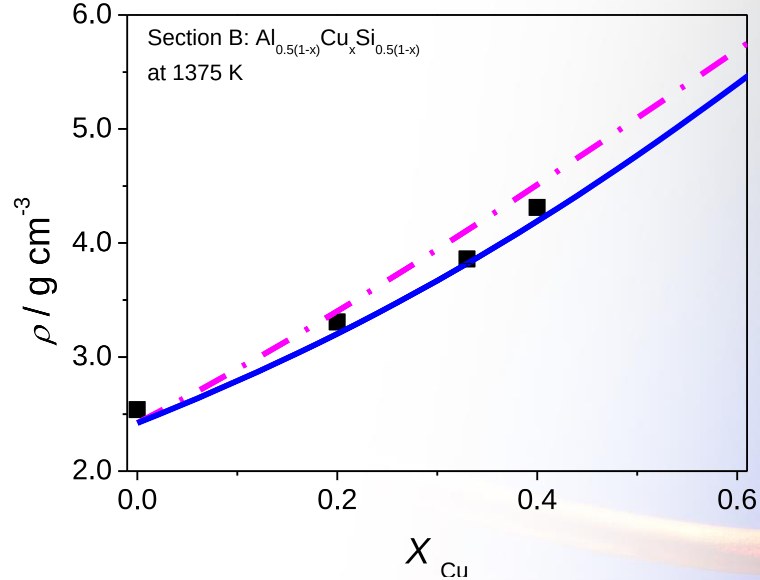
For binary systems: $V^{Al, Cu}$ = -3.87 [1)] , V^{Cu-Si} = -2,38 [2)], V^{Al-Si}= 0 [3)],
(unit: $cm^3 mol^{-1}$)

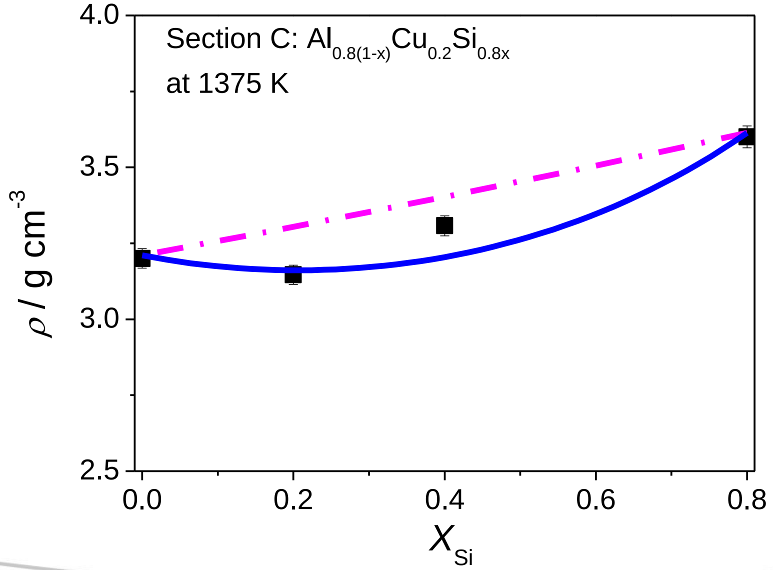
For ternary Al-Cu-Si = ?

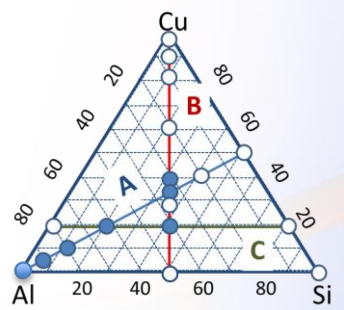
1) Brillo, Egry. Westphal, Int. J. Mat. (2008), 2) Adachi, Schick, Brillo, Egry Watanabe, J. Mater. Sci. (2010)
3) Brillo Egry , Jap J. Apple Phy. (2011)

Al-Cu-Si – isothermal density









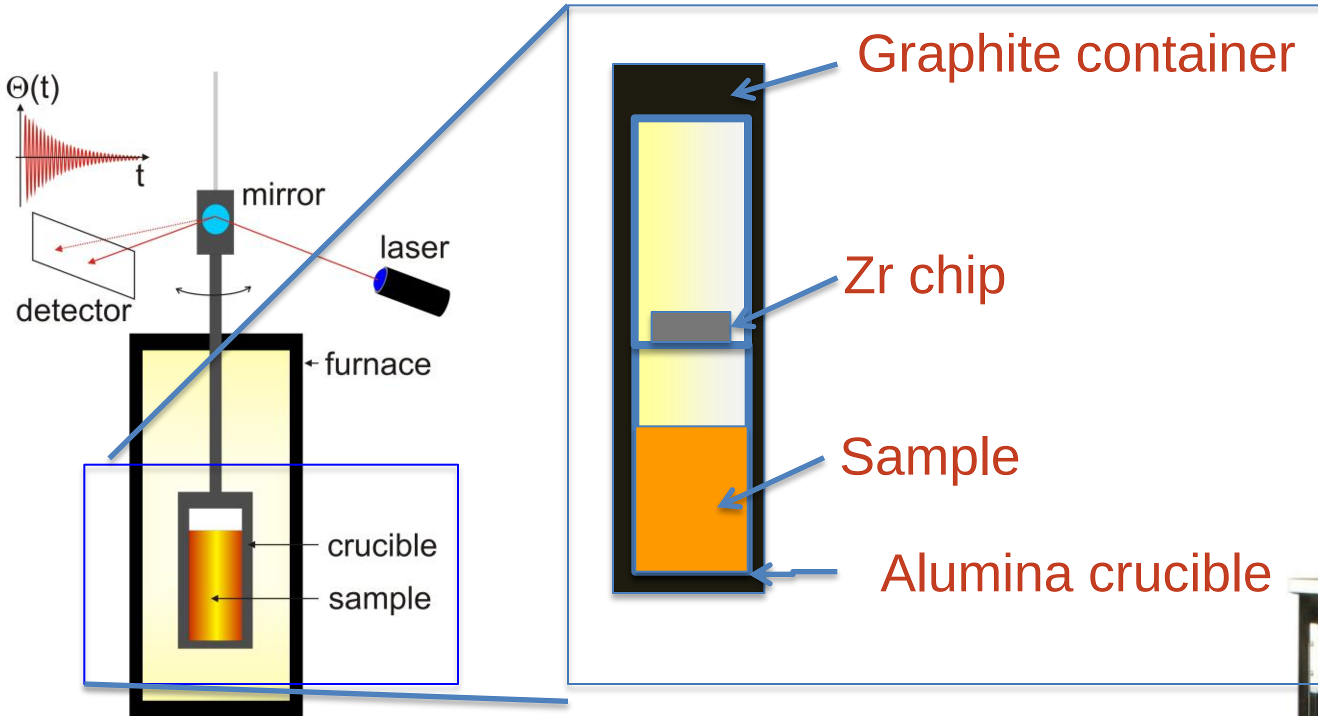
^{T}V= 19.7 cm^{3}mol^{-1}

^{T}V= 0

For ternary Al-Cu-Si: ^{T}V = 19.7

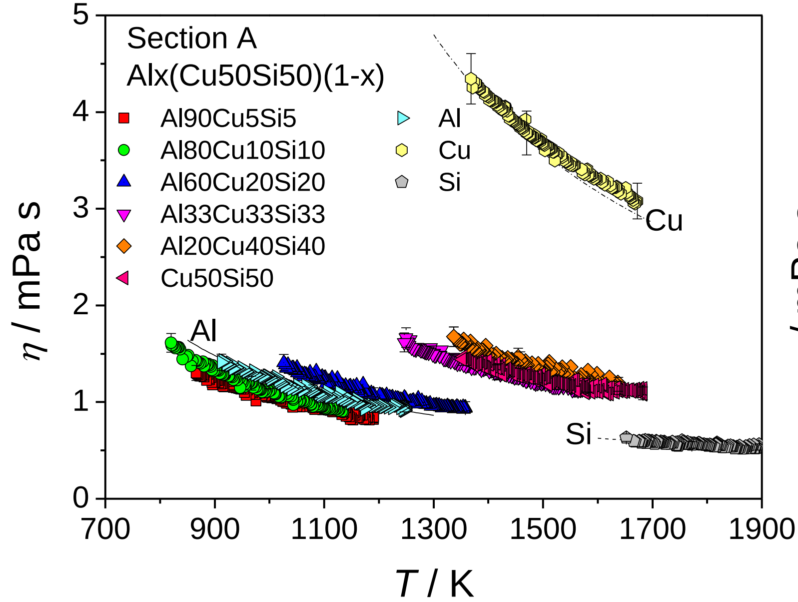
Viscosity measurement

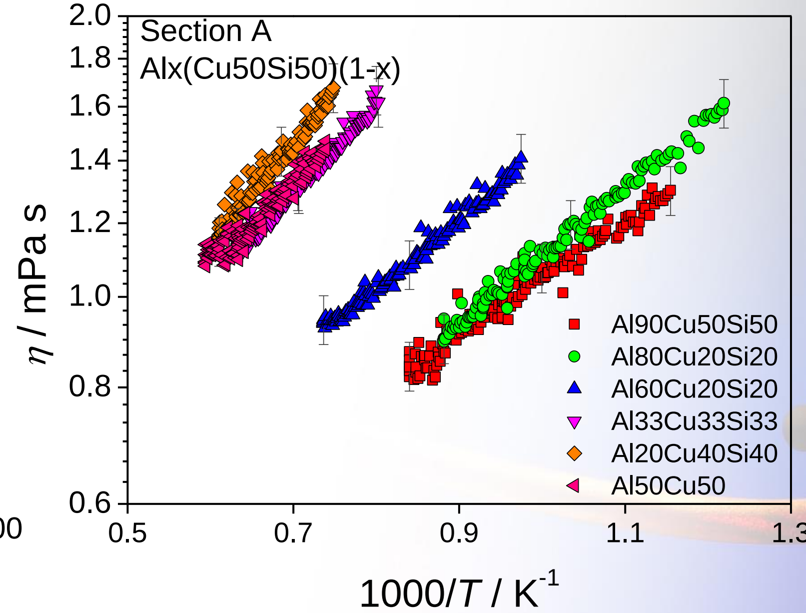
oscillating cup method



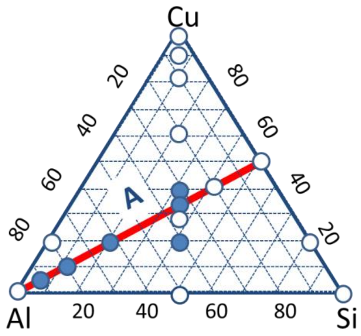
- measurements up to 2300 K
- smooth continuos curves are obtained
- crucible – melt interaction must be considered

Viscosity of liquid Al-Cu-Si





Al: Brillo et al. (2007), Cu: Iida and Guthrie (1993), Si: Sato et al. (2003)



Precise data for entire composition range

Broad T-range: $T_L < T < 1900$ K

Viscosity can be expressed with Arrhenius form

(Arrhenius: $\ln(\eta) = \ln(\eta_\infty) + E/RT$)

Models for the viscosity of liquid alloys

Kaptay [1)]

$$\eta = \frac{hN_A}{V}\exp\left(\frac{\sum_i x_i \cdot \Delta E_{A,i} - 0.155\Delta H_{mix}}{RT}\right)$$

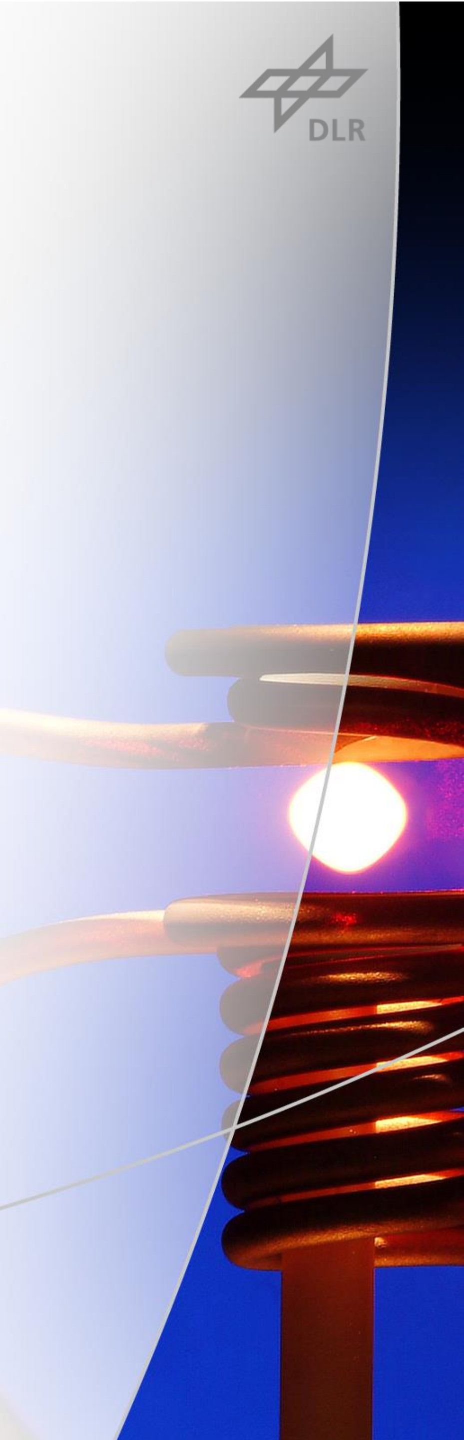
Kozlov et al. [2)]

$$\ln\eta = \sum_{i=1}^{N} x_i \ln\eta_{i,\infty} + \frac{\sum_{i=1}^{N} x_i \cdot E_i - \Delta H_{mix}/3}{RT}$$

Schick et al. [3)]

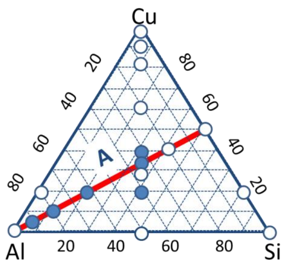
$$\ln\eta = \sum_{i=1}^{N} x_i \ln x_i\eta_{i,\infty} + \frac{\sum_{i}^{N} x_i \cdot E_i - \Delta H_{mix}}{RT}$$

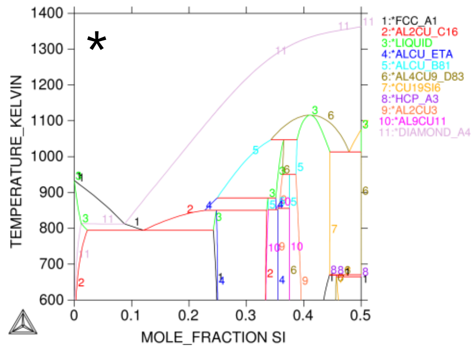
Which model can explain the experimental results?

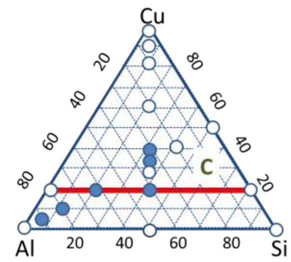


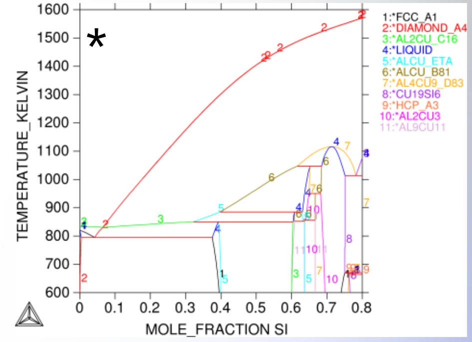
1), Kaptay. Proc. micro CAD (2003), 2) Kozlov. Cher. Meya (1983),
3) Schick, Brillo, Hallstedt, Egry, J. Mat. Sci. (2012)

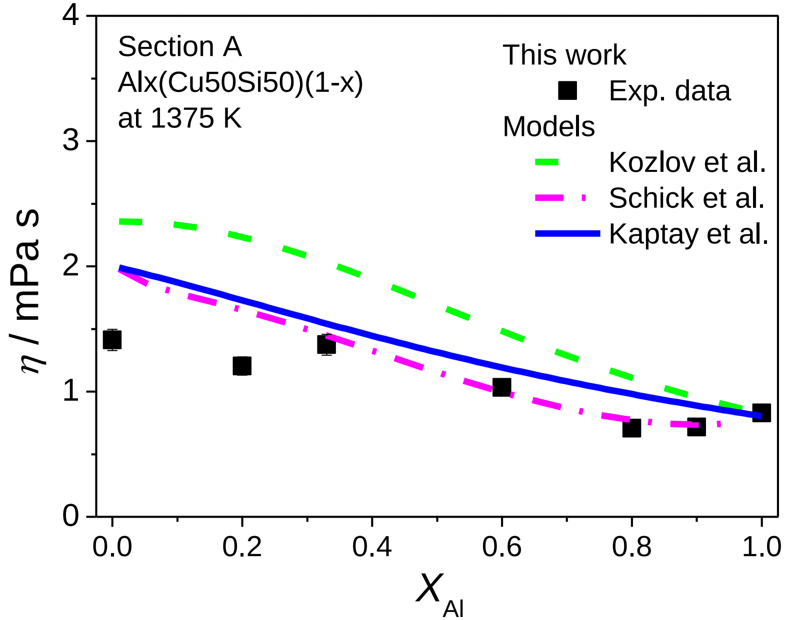
Viscosity of liquid Al-Cu-Si

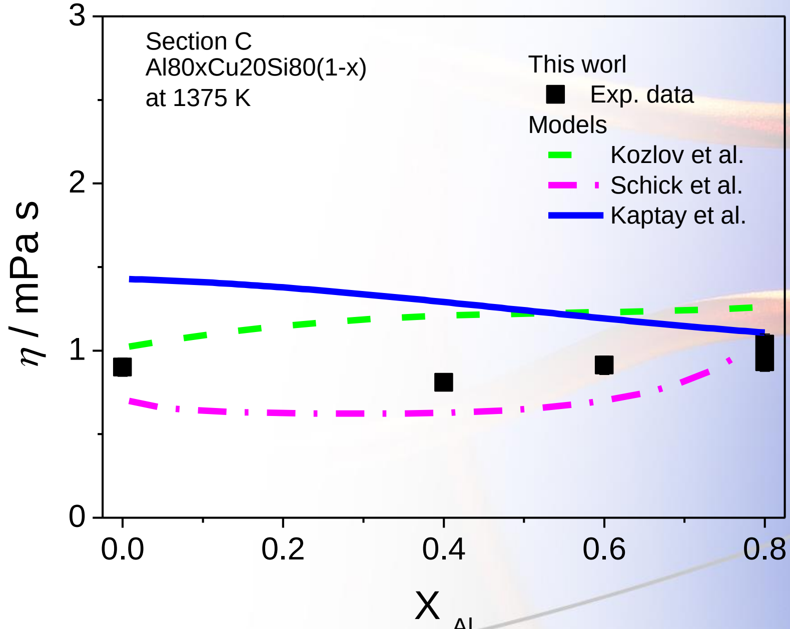






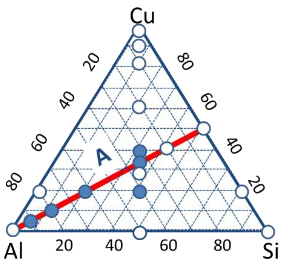


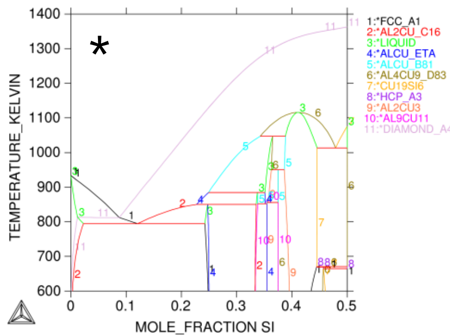


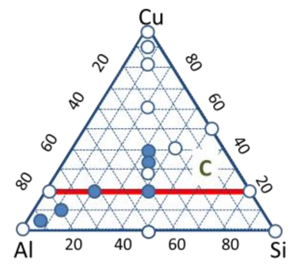


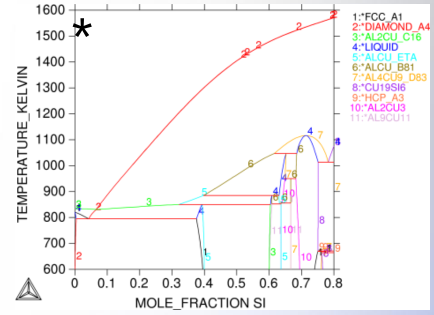
* Calculated by Dr. Bengt Hallstedt

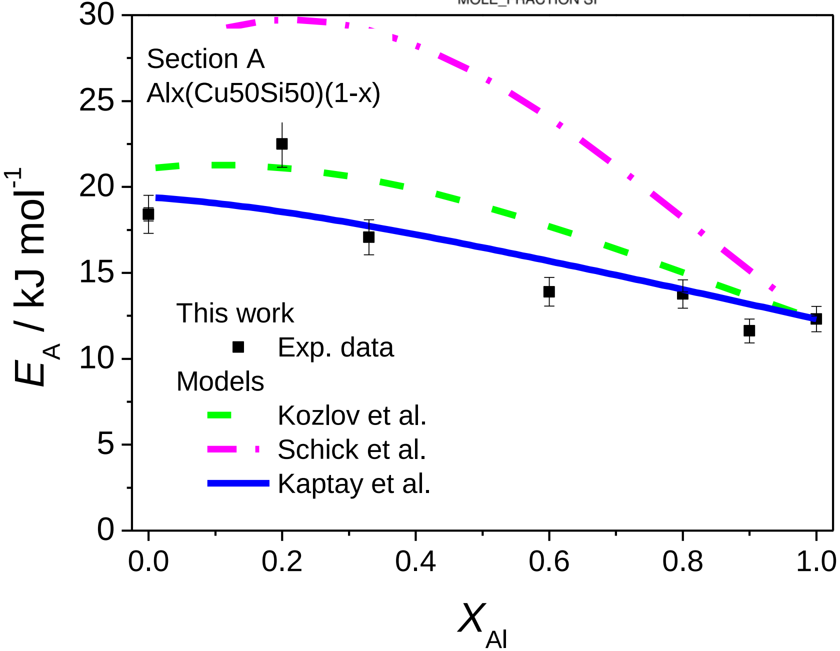
Activation energy of viscos flow for Al-Cu-Si liquid

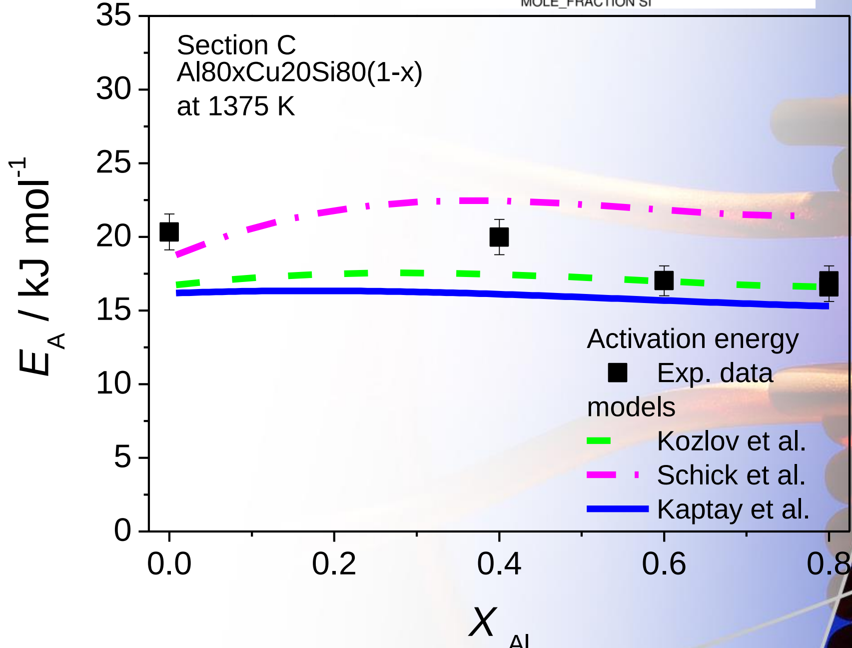








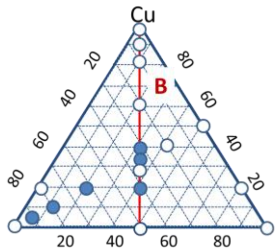


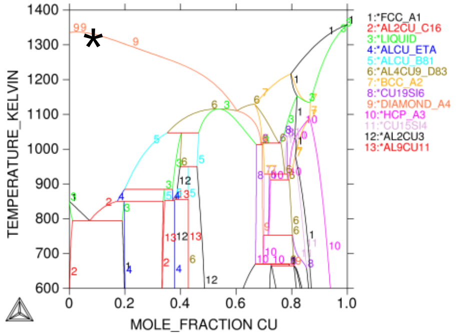


Kaptay model well explains the viscosity along section A and C

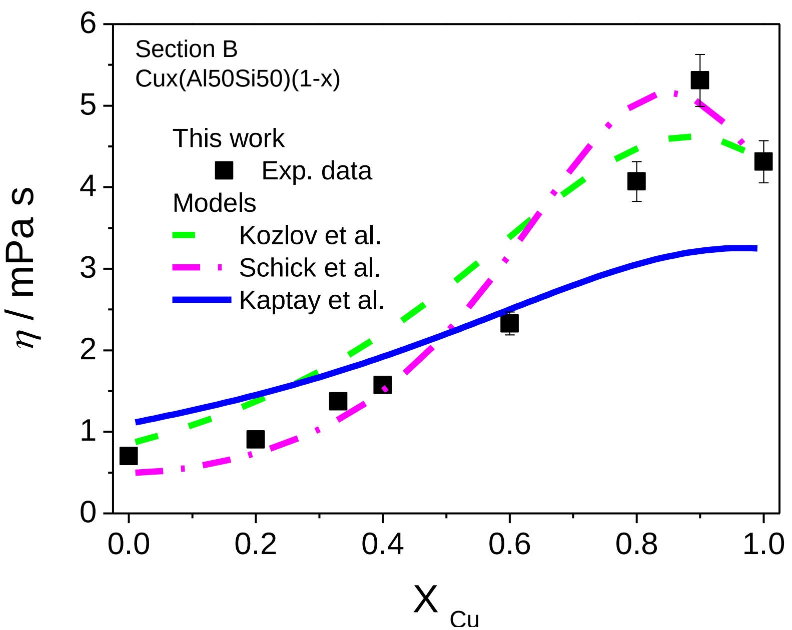
$$\eta = \frac{hN_A}{V}\exp\left(\frac{\sum_i x_i \cdot \Delta E_{A,i} - \alpha \Delta H_{mix}}{RT}\right)$$

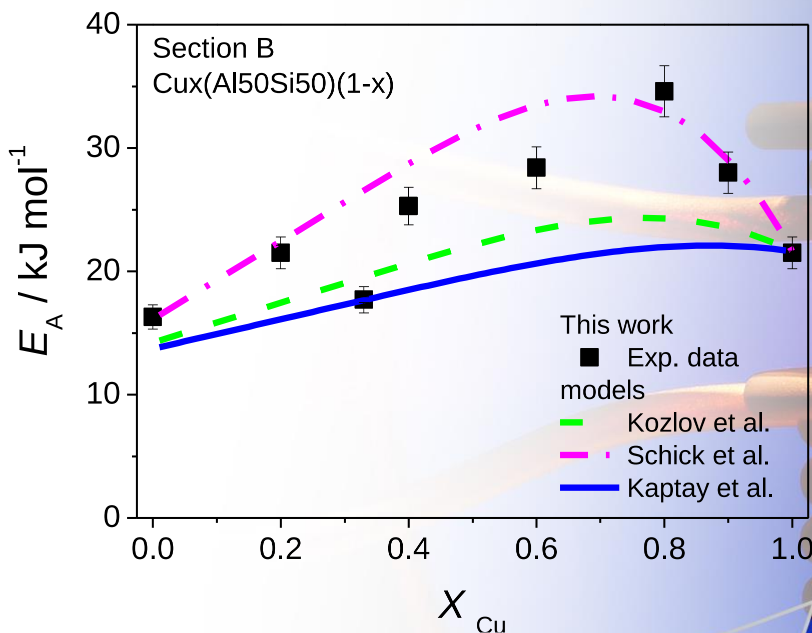
Viscosity and activation energy of viscous flow of liquid Al-Cu-Si





* Calculated by Dr. Bengt





Schick model well explains the viscosity along section B

$$\ln\eta = \sum_{i=1}^{N} x_i \ln x_i \eta_{i,\infty} + \frac{\sum_{i}^{N} x_i \cdot E_i - \Delta H_{mix}}{RT}$$

Summary

Density of Al-Cu-Si ternary liquid alloys:

- Density has been measured over broad T-range
- Ternary interaction parameter was determined as

  $^{T}V = 19.7\ \mathrm{cm^3\ mol^{-1}}$.

Viscosity of Al-Cu-Si ternary liquid alloys:

- Viscosities measured for entire concentration and broad T-range

- Viscosity can be described by
  1) Kaptay model for eutectic system
  2) Schick model for CFM like system

Thank you for your attention

Thermophysical properties of Al-Si-Mg-Cu melt
funding by DFG under contract BR 3665/3-1