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Surface tension and density of liquid Ag-Cu alloys

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Presentation plan

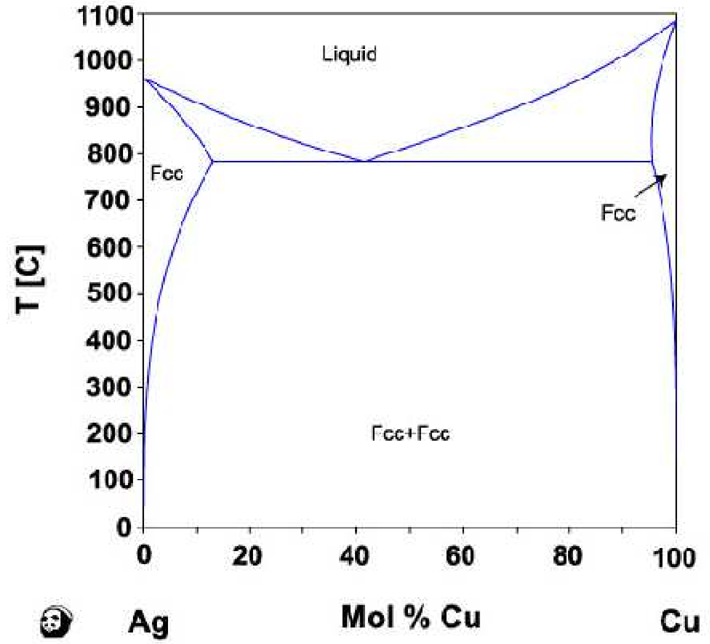
- Introduction
- Results for pure Ag
- Results for Ag-Cu alloys
  - Density and molar volume
  - Surface tension
  - Modelling of the surface tension
- Summary

Introduction

Ag-Cu system:

- A subsystem of ternary and multicomponent solders
  - Ag-Cu-Sn
  - Ag-Cu-Sn-In

- A braze alloy for joining of metals and ceramics
  - Ag-Cu
  - Ag-Cu-Ti



www.nims.go.jp/cmsc/pst/database/agelem/agcu/

Introduction

- **Investigated by a few groups:**
  - **relatively good agreement of density data**
  - **some differences in surface tension data**

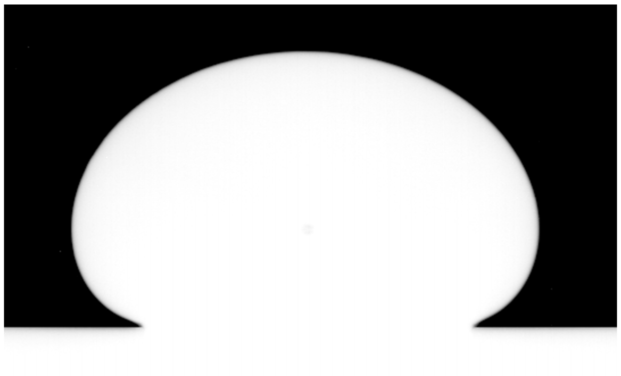
Author (Year)	Density	Surface tension
Krause et al. (1929)	+	+
Bricard et al. (1973)		+
Sebo et al. (1977)	+	+
Lee et al. (2004)		+
Novakovic et al. (2005)		+
Kucharski et al. (2006)	+	+
Brillo et al. (2006)	+	

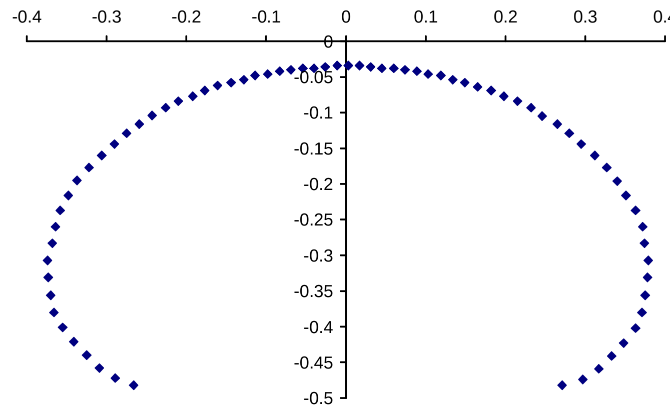
Experimental procedure

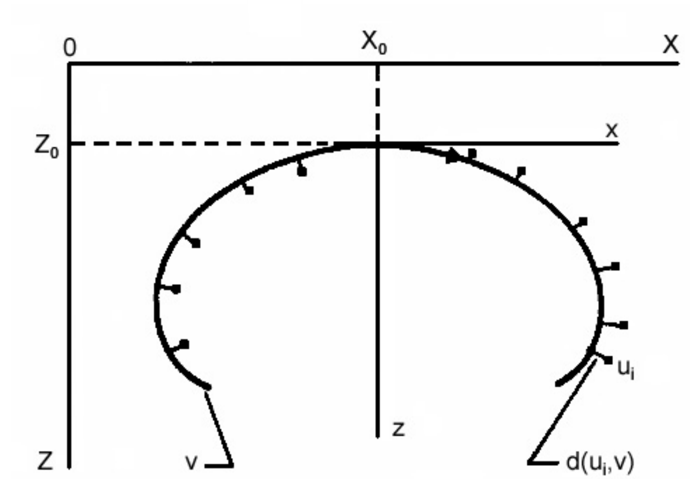
- **Materials:**
  Ag (5N), X(Cu): 0.3; 0.4; 0.5
- **Sessile drop method**
- **Ar (6N) protective gas**
- **1098 – 1573 K, temperature decreasing stepwise**
- **Hold time 10 min**
- **Surface tension calculated with AstraView®, developed by CNR-IENI, Genoa, Italy**

N. Sobczak et al. Mater Sci Eng A495 (2008) 43

Surface tension measurement







1. capture the image
2. change the image into a set of coordinates
3. fit the solution of Laplace equation to coordinates

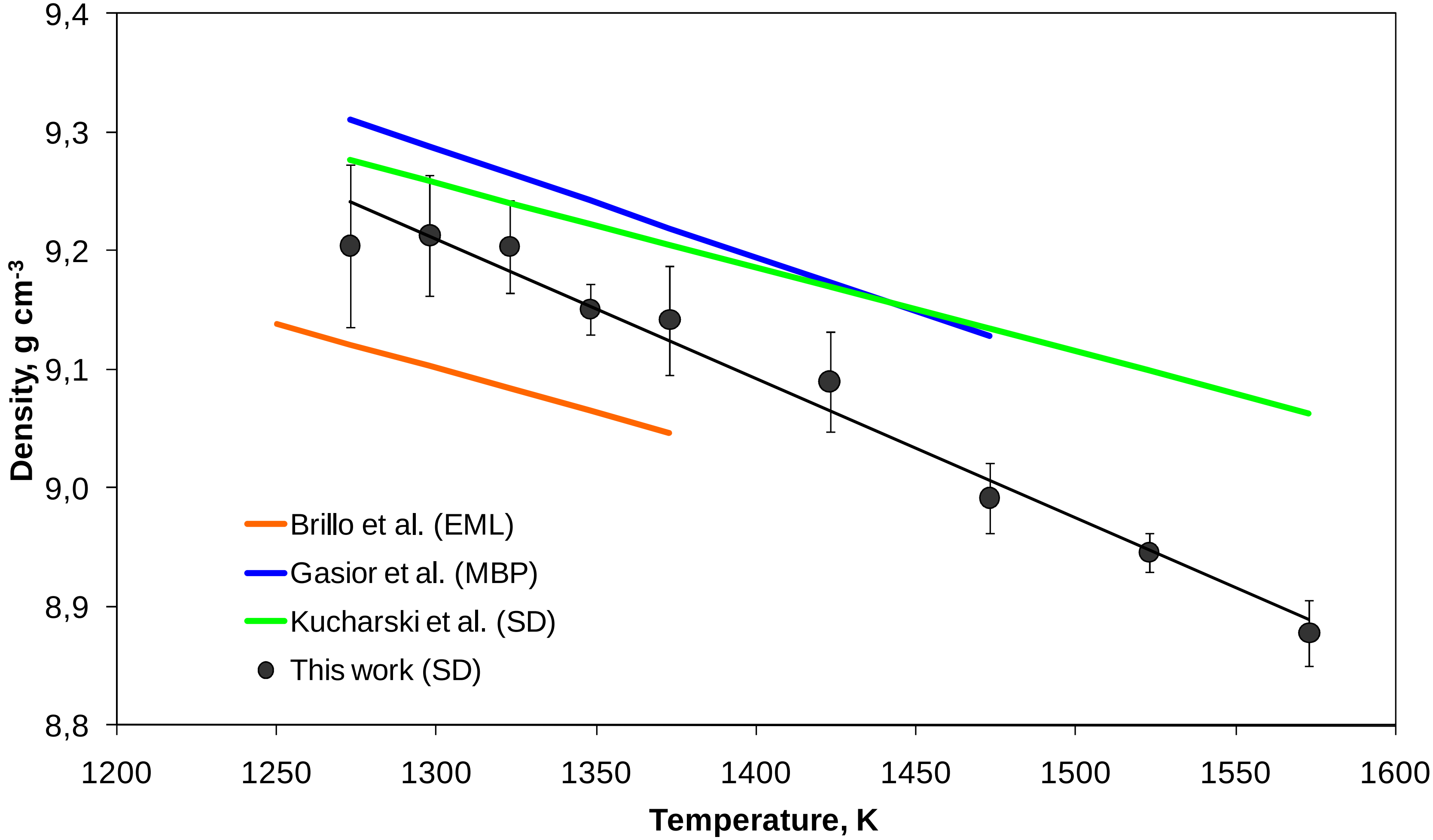
Rotenberg et al., J Colloid Interf Sci 93 (1983) 169

Ag (pure) - density

ρ_L (g· cm^{-3})	ρ_T (g·cm^{-3} K^{-1})	Method	Atmosphere	Temperature (K)	Year
9.237	-0.00065	A	–	1273 – 1473	1929
9.28	-0.0009	A	–	1234 – 1573	1951
9.302	-0.000959	A	N_2	1234 – 1573	1953
9.348	-0.0011	MBP	H_2	1323 – 1423	1960
9.346	-0.0009067	A	–	1234 – 2450	1962
9.337	-0.00105	MBP	Ar	1234 – 1673	1964
9.36	-0.00108	MBP	Ar	1273 – 1473	1969
9.301	-0.00105	SD	Ar	1253 – 1381	1971
9.32	-0.00097	A	N_2 + 10% H_2	1234 – 1773	1972
9.321	-0.0009787	A	–	1234 – 1400	1975
9.31	-0.00078	SD	Ar, H_2	1234 – 1873	1989
9.346	-0.000911	MBP	Ar + 10% H_2	1273 – 1473	2003
9.304	-0.00071	SD	Ar	1273 – 1523	2005
9.15	-0.00074	EML	He + 8% H_2	1250 – 1380	2006
9.288	-0.001175	SD	Ar	1273 – 1573	2010

A – Archimedean principle, EML – electromagnetic levitation, MBP – maximum bubble pressure, SD – sessile drop method

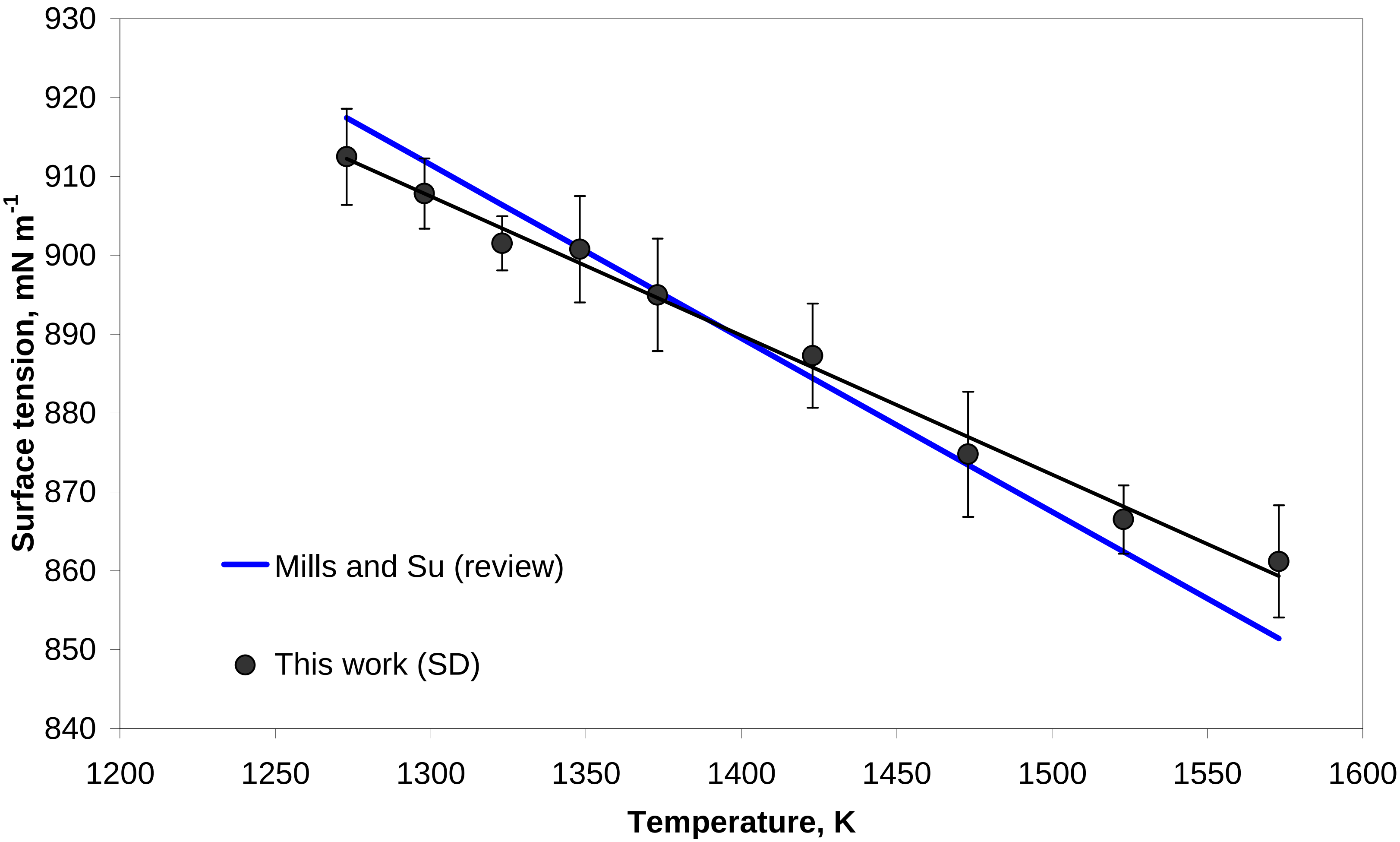
Ag (pure) - density



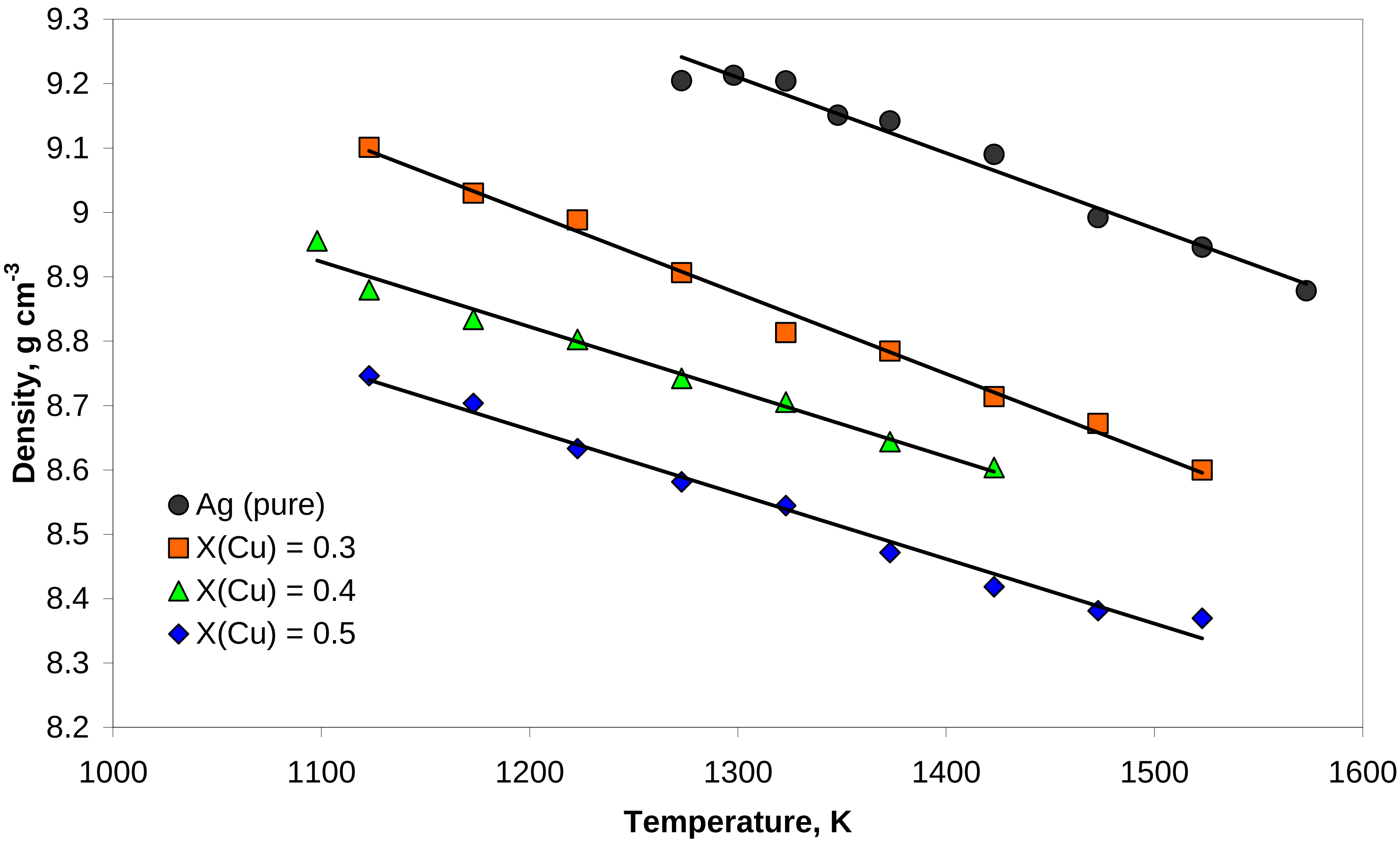
Ag (pure) – surface tension

ρ_L (mN· m^{-1})	σ_T (mN·m^{-1} K^{-1})	Method	Atmosphere	Temperature (K)	Year
946.0	-0.4	SD	Vac.	1248 - 1298	1953
884.9	-0.17	MBP	-	1273 - 1473	1958
898.4	-0.135	MBP	-	1273 - 1473	1958
900.4	-0.1883	MBP	H_2	1273 - 1523	1960
913.0	-0.155	MBP	H_2	1273 - 1473	1964
921.3	-0.174	MBP	-	1800 - 2330	1968
957.5	-0.14	SD	Vac.	1246 - 1346	1970
910.7	-0.117	SD	Ar	1253 - 1381	1971
926.0	-0.15	SD	He	1234 - 1573	1972
890.0	-0.16	SD	He	1253 - 1373	1982
910.0	-0.17	SD	Ar, H_2	1270 - 1860	1986
924.0	-0.19	EML	Ar, H_2	1273 - 1383	1986
898.3	-0.1905	MBP	Ar + 10% H_2	1273 - 1473	2003
925.6	-0.228	SD	Ar + 10% H_2	1269 - 1473	2004
914.0	-0.15020	SD	Ar	1234 - 1473	2005
912.3	-0.204	SD	Ar	1273 – 1523	2005
984	-0.298	EML	Ar + 5% H_2	1234 – 1734	2008
919.8	-0.176	SD	Ar	1273 – 1573	2010

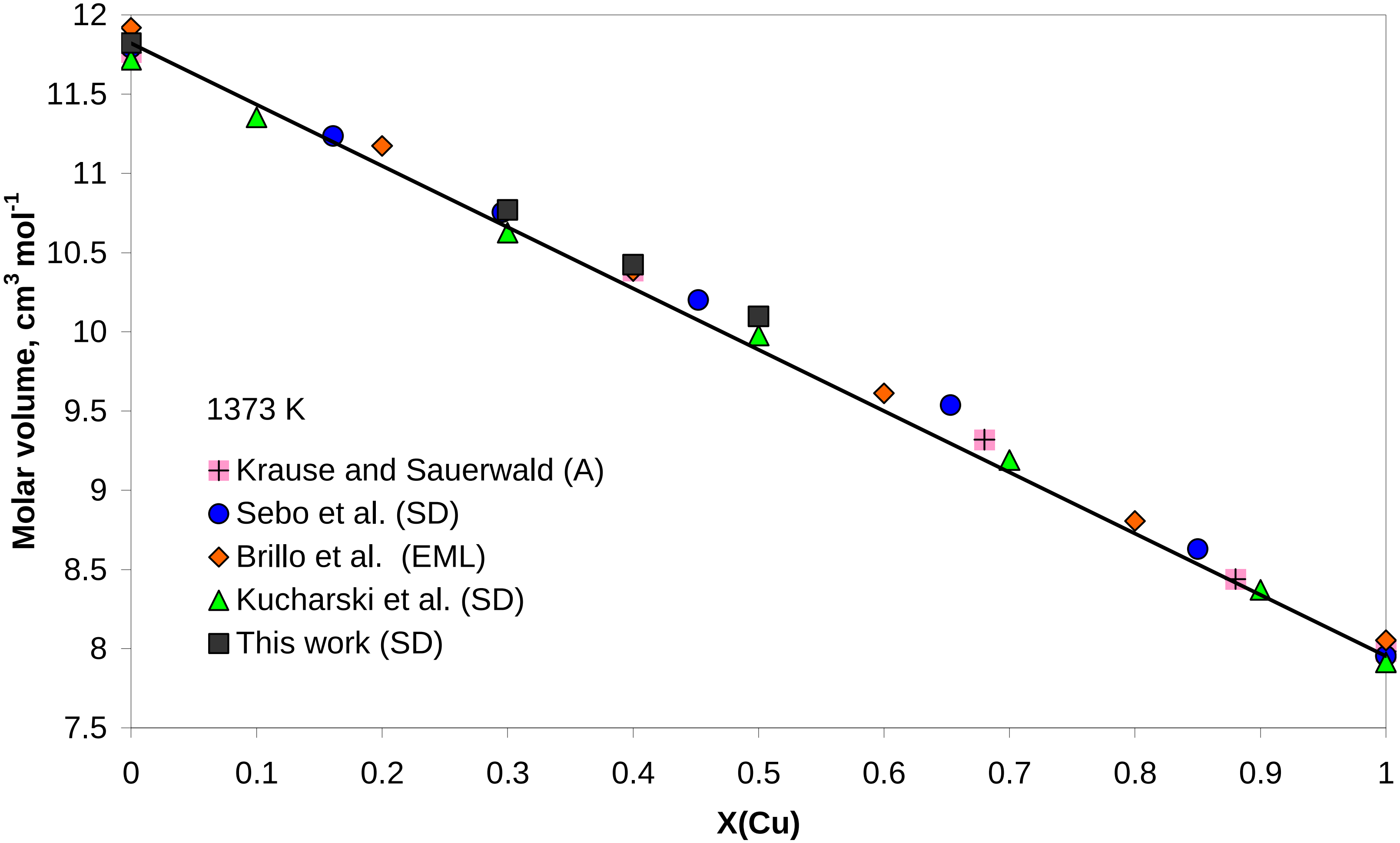
Ag (pure) – surface tension



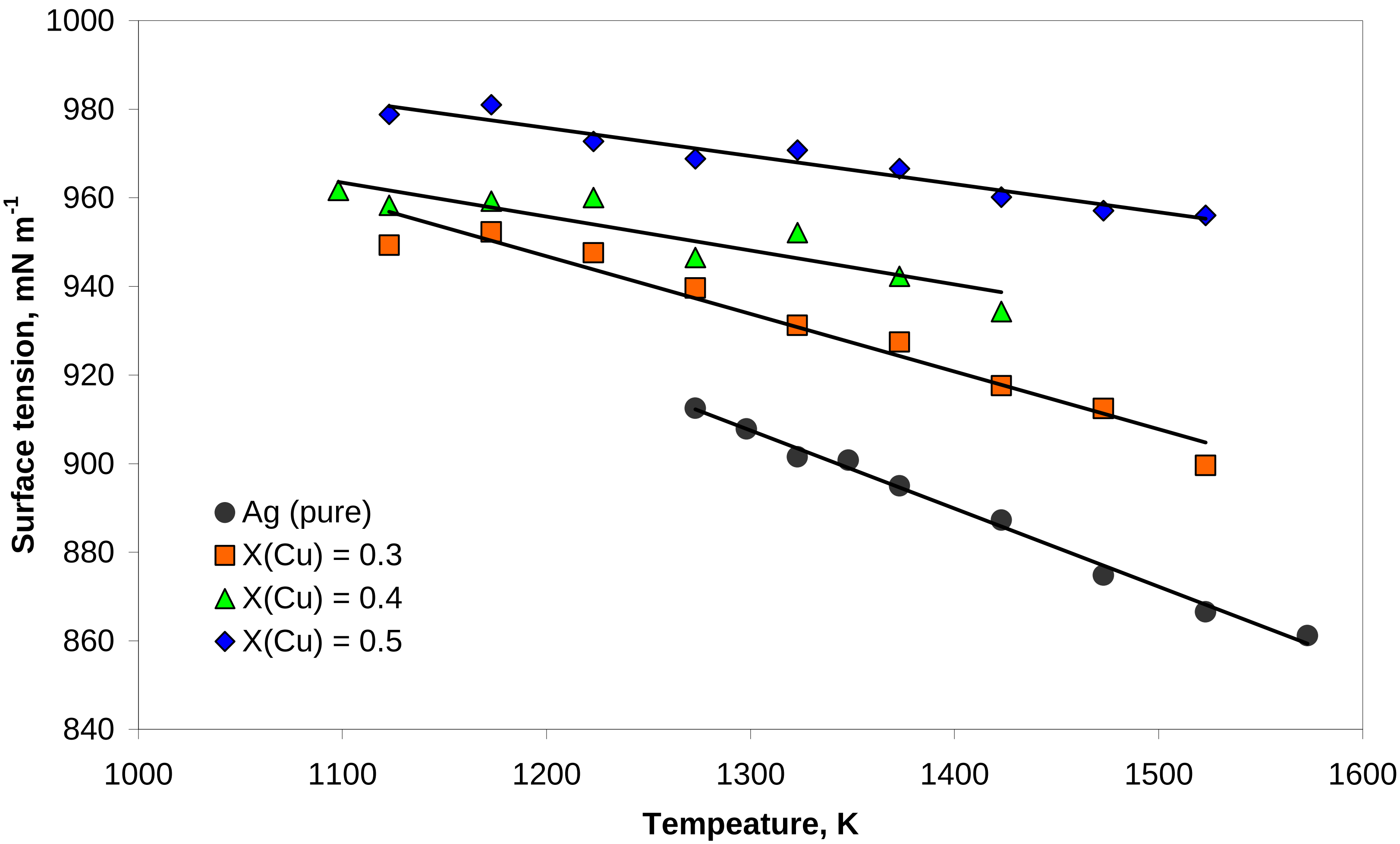
Ag-Cu alloys - density



Ag-Cu alloys – molar volume



Ag-Cu alloys – surface tension



Modelling of the surface tension

- **The Butler equation**

$$\sigma = \sigma_i + \frac{RT}{A_i} \ln \frac{x_i^{(s)}}{x_i} - \frac{1}{A_i} \left(G_i^{Ex(s)} - G_i^{Ex} \right)$$

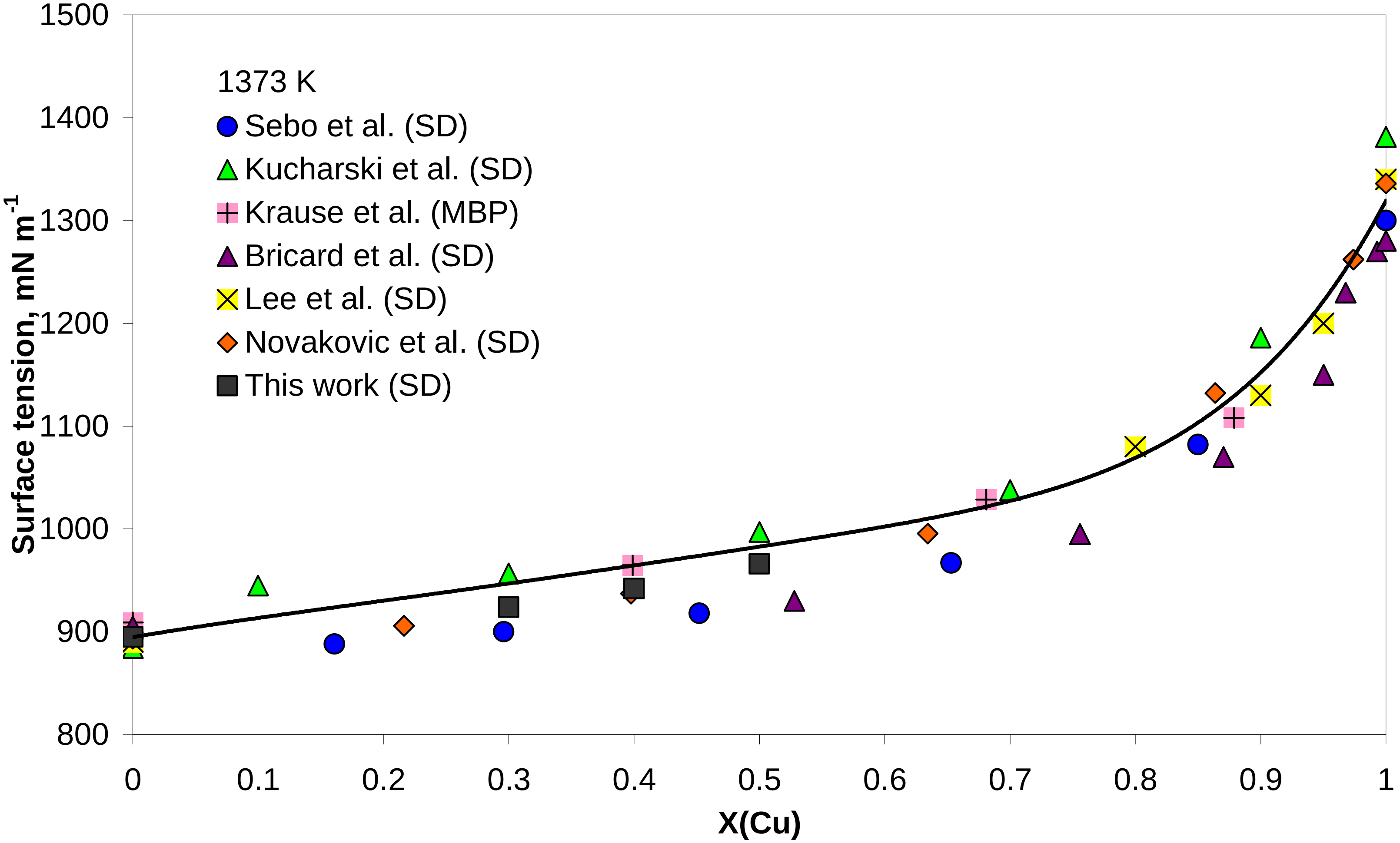
- **The partial Gibbs free energy of the component *i***

$$G_i^{Ex} = (x_j)^2 \left[L_{i-j}^{(0)} + L_{i-j}^{(1)} (x_j - 3x_i) + L_{i-j}^{(2)} (x_j - x_i)(x_j - 5x_i) + \ldots \right]$$

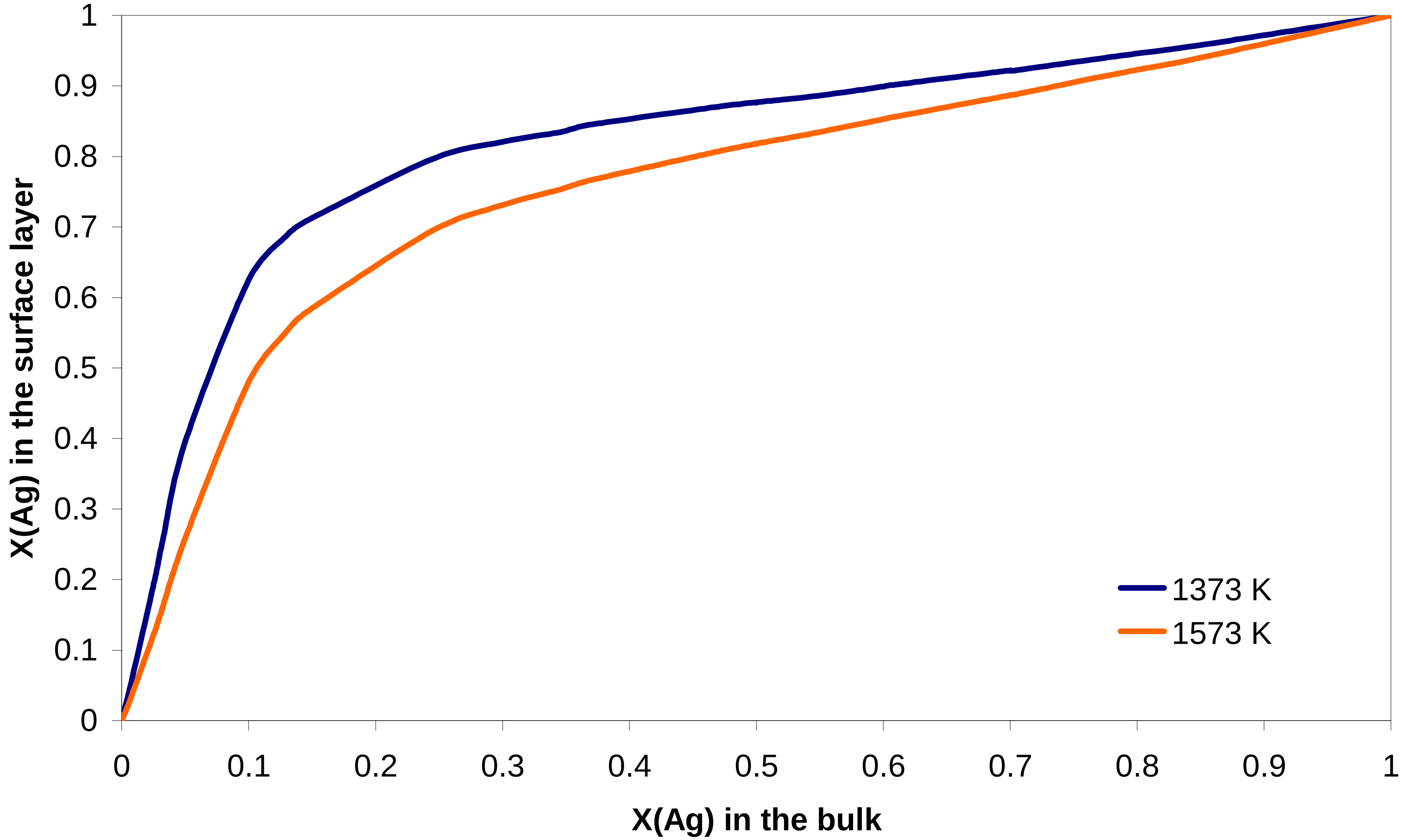
$$G_i^{Ex(s)} = \beta \cdot G_i^{Ex}$$

- **Parameters *L* from COST 531 thermodynamic database**

Modelling of the surface tension



Modelling of the surface tension



Summary

- **Our data for pure Ag agree well with literature data**
- **Density of Ag-Cu alloys decreases with increasing temperature and with increasing concentration of Cu**
- **Molar volume shows additive behavior in the whole range of Ag-Cu compositions**
- **Surface tension decreases with temperature and increases with Cu content, a small change of slope is observed**
- **There is a good agreement of experimental data with surface tension calculated with the Butler equation**
- **Surface layer of Ag-Cu solution is enriched with silver**

P. Fima, N. Sobczak (2010) Int J Thermophys, doi: 10.1007/s10765-010-0798-5

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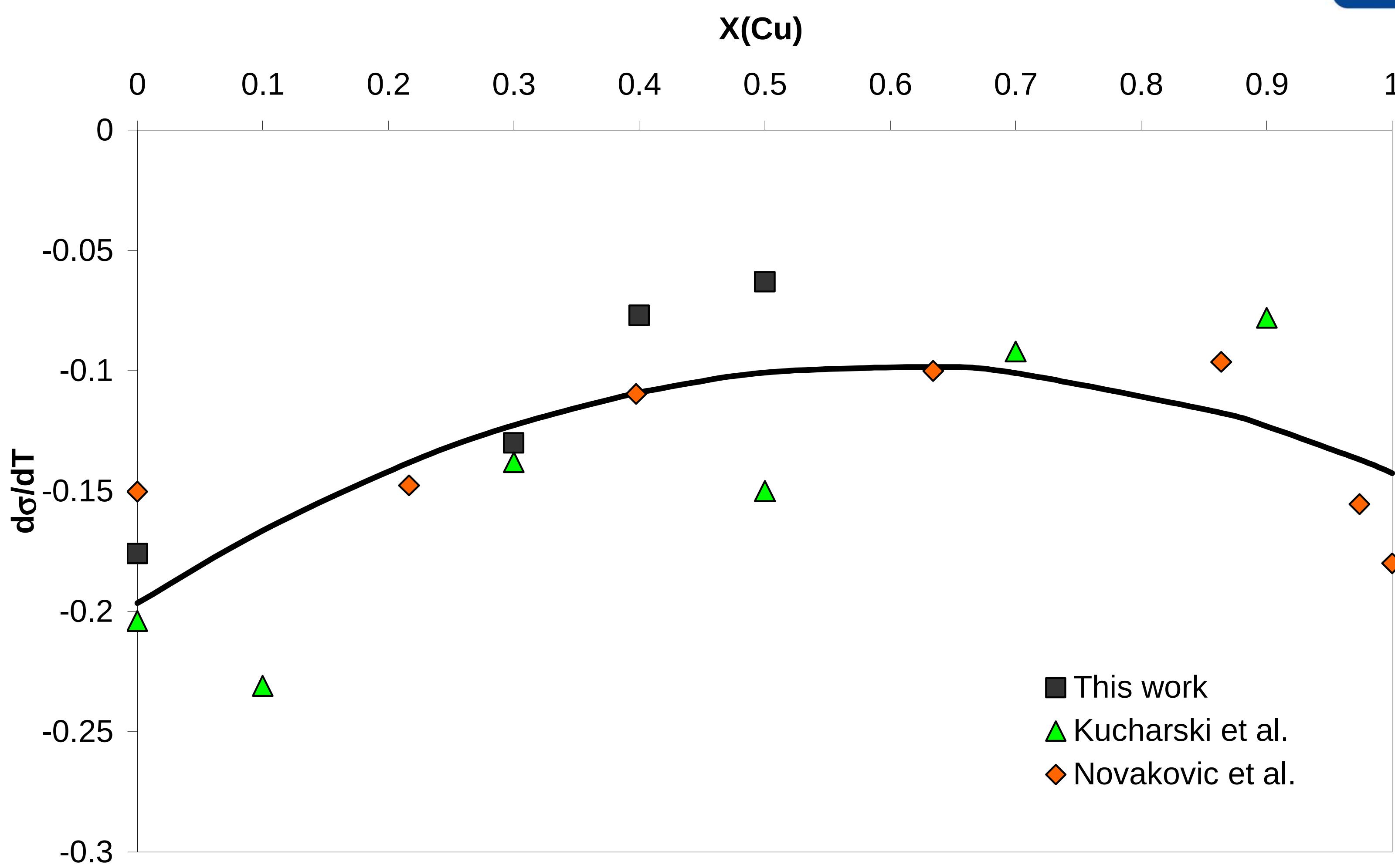
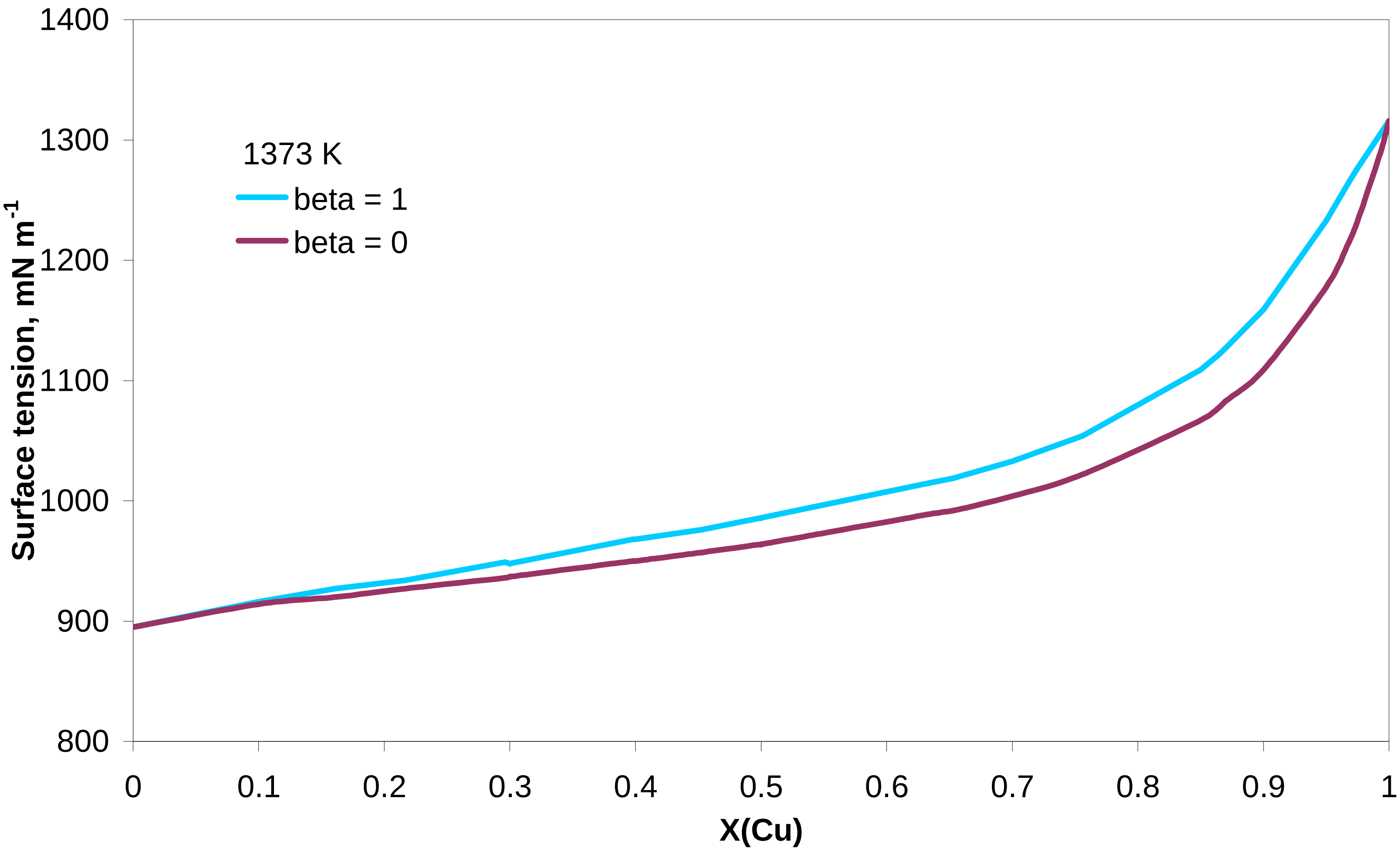
Thank you for your attention!

Ag-Cu alloys – surface tension



Modelling of the surface tension



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$$\sigma\left(\frac{1}{r_1}+\frac{1}{r_2}\right)=\rho g\left(h-z\right)+C$$