On the thermochemical modelling of viscosities of liquid oxide mixtures

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Structure of presentation

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- I.0. Introduction
- I.1. Effect of data sorting
- I.2. Effect of temperature
- I.3. Effect of composition
- I.4. Summary

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- II.0. Introduction
- II.1. Structure of oxide mixtures
- II.2. Associate species thermodynamic model
- II.3. Avramov equation
- II.4. Viscosities of binary systems
- II.5. Temperature dependence
- II.6. Summary

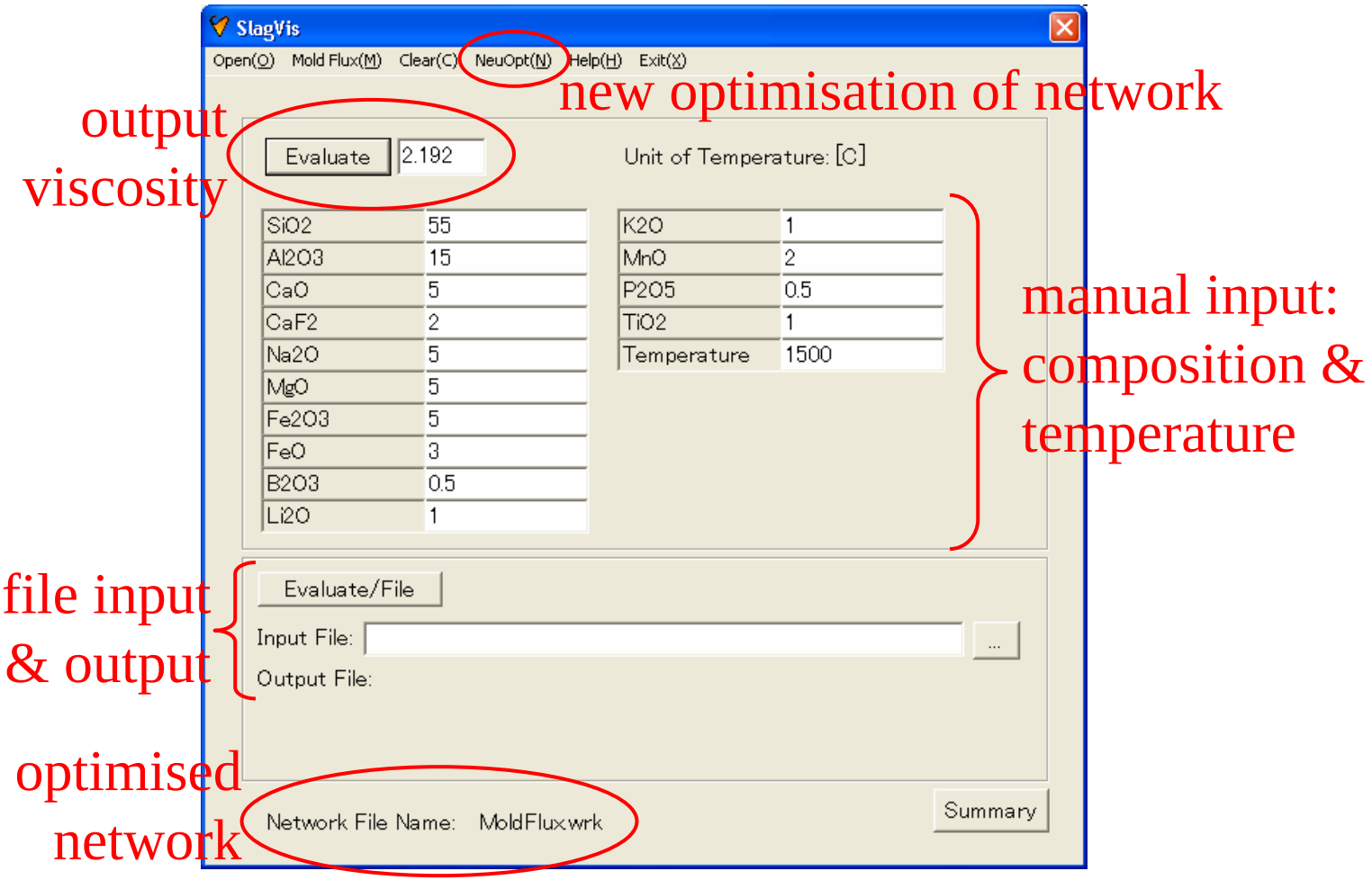
III. Modelling heterogeneous mixtures

I. Application of neural networks

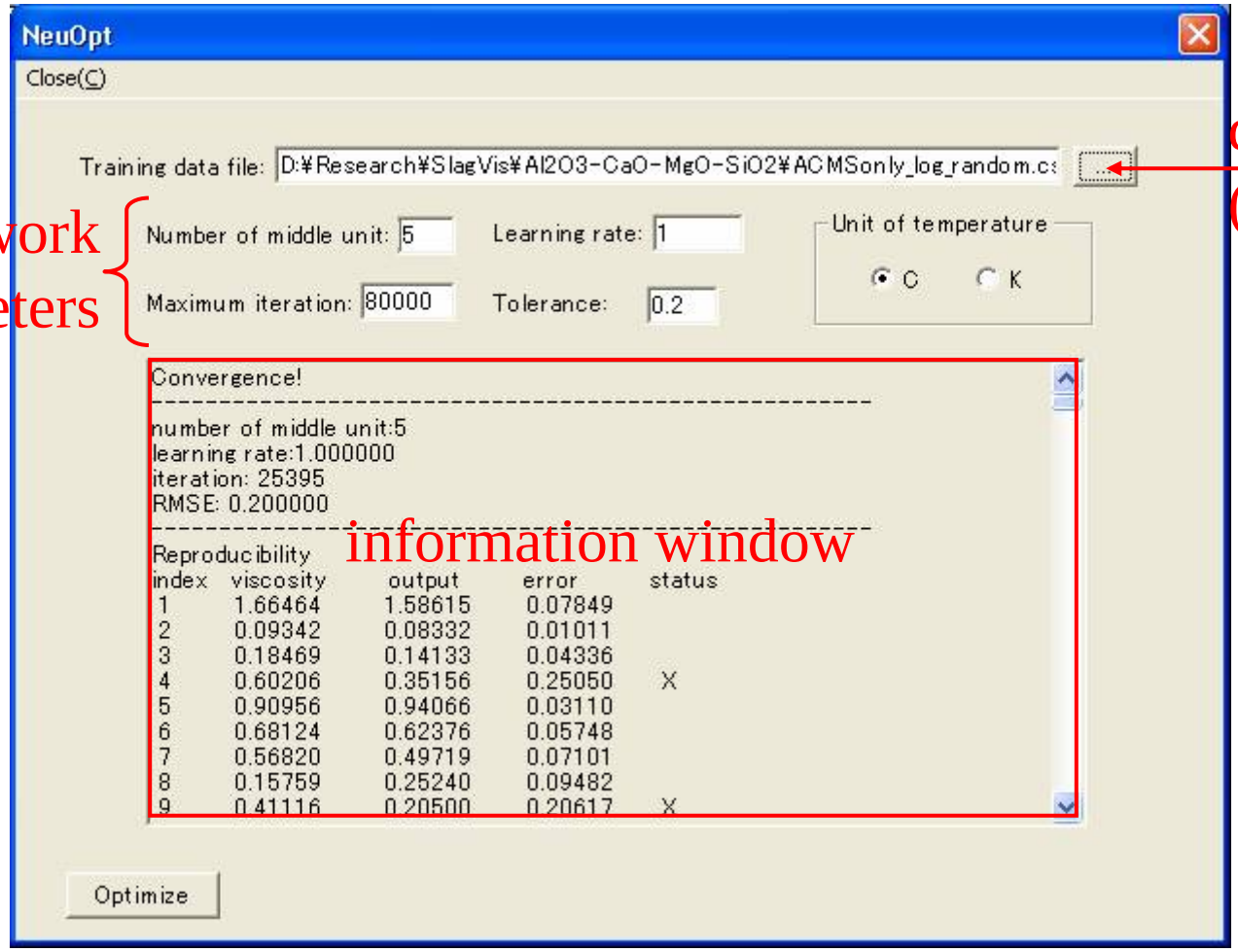
I.0. Introduction – SlagVis

- SlagVis (Research Centre of Computational Mechanics, Japan)
- Based on neural network model (Hanao, Kawamoto, Tanaka, Nakamoto, 2006)
- Learning algorithm: sequential backpropagation method

I.0. Introduction – SlagVis



I.0. Introduction – SlagVis



network parameters

dataset (csv-file)

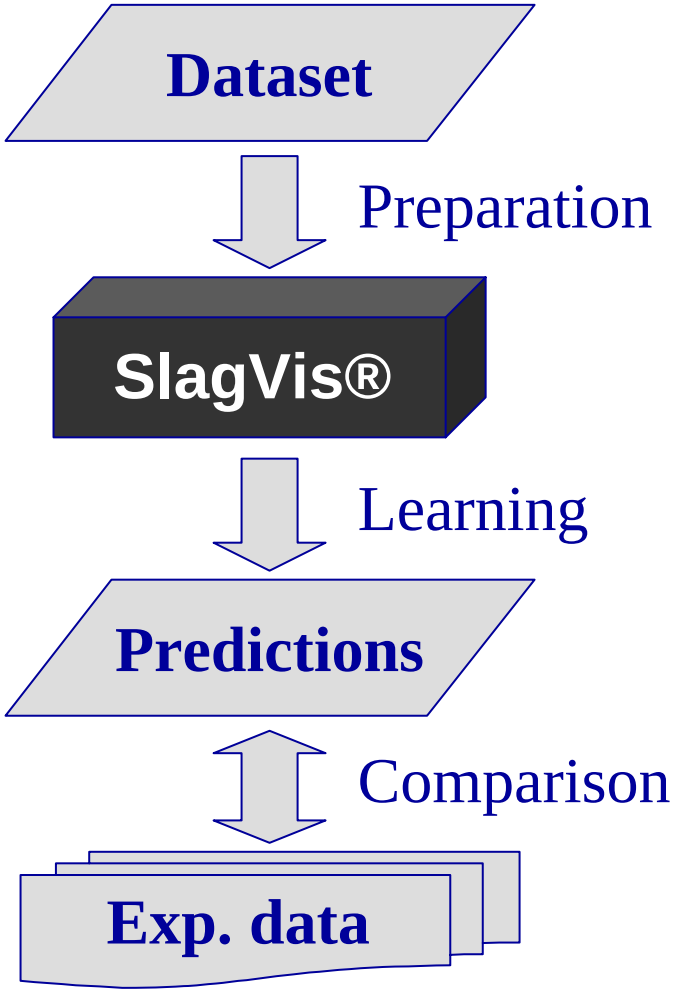
information window

I.0. Introduction – viscosity database

Viscosity database (Kondratiev, 2004-2008):

- > 18,500 experimental points
- > 2,500 compositions (synthetic systems)
- Al_2O_3, CaO, FeO, Fe_2O_3, K_2O, MgO, Na_2O, PbO, $SiO_2 + B_2O_3$, BaO, GeO_2, Li_2O, MnO, SrO
- T range: 300 – 2500^oC

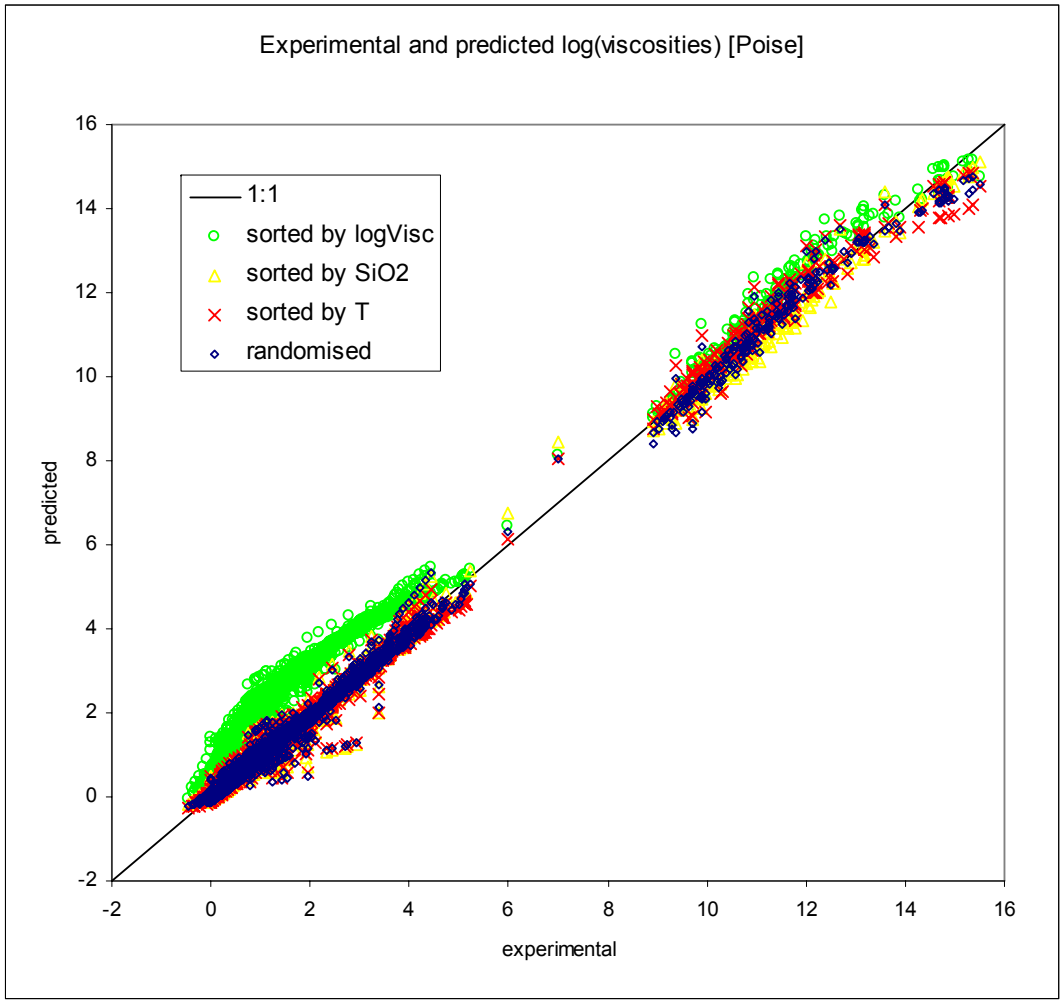
I.0. Introduction - scheme of tests



Tests carried out:

- Data sorting
- Temperature inter- and extrapolation
- Composition inter- and extrapolation

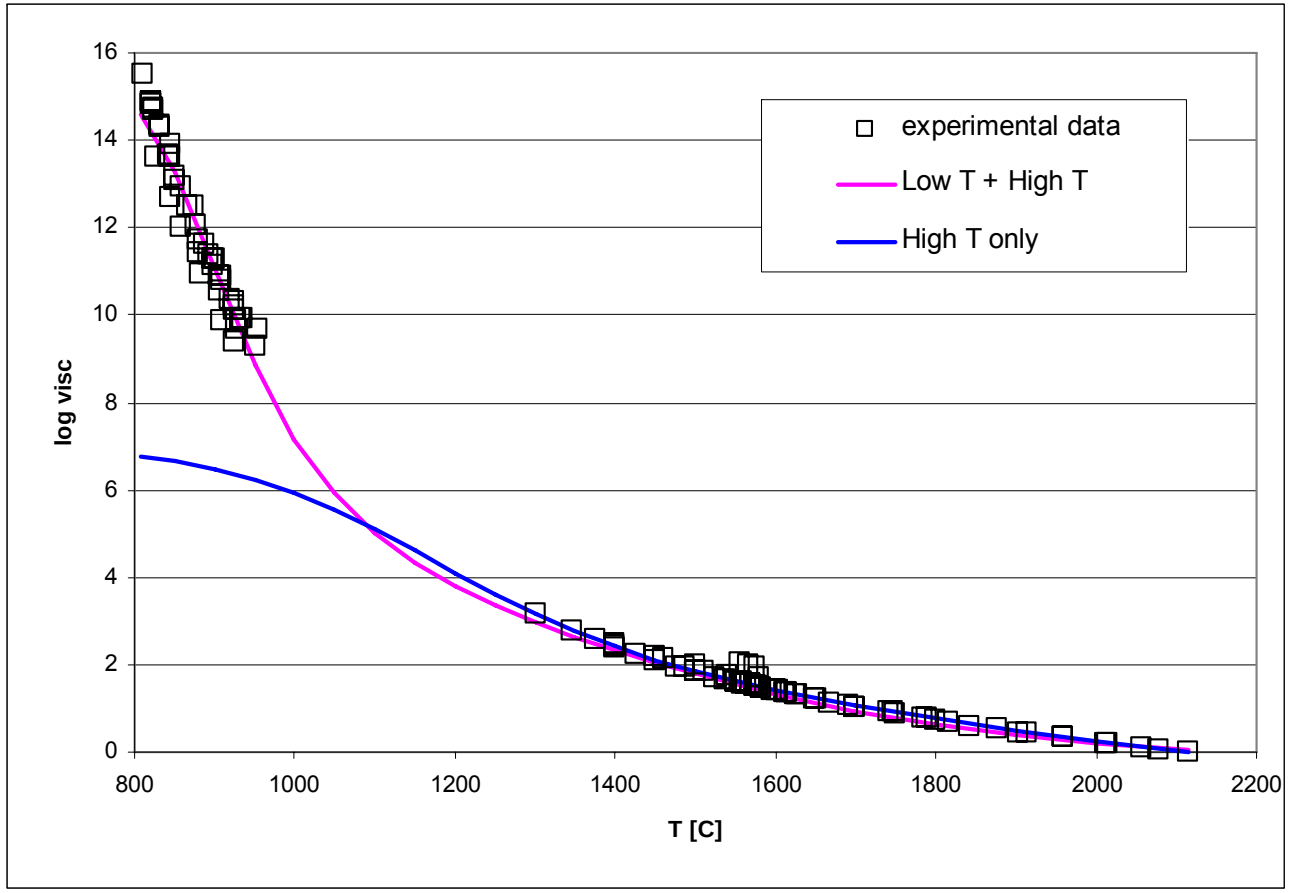
I.1. Effect of data sorting



- Sequential backpropagation algorithm depends on data sorting
- Randomisation necessary
- Batch backpropagation algorithm being implemented

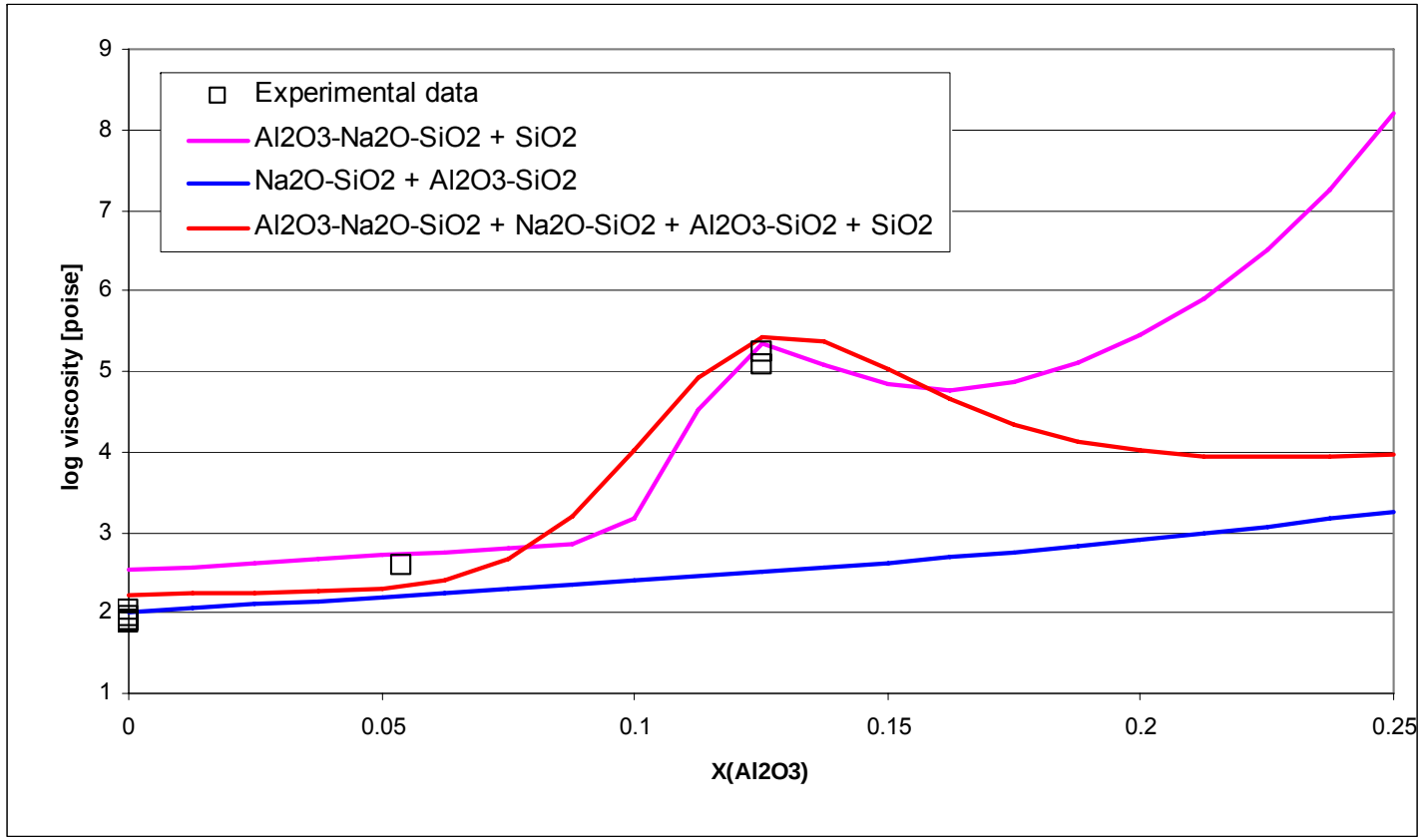
I.2. Effect of temperature

Al_2O_3-CaO-SiO_2, X_{SiO2}=0.5, X_{Al2O3}=X_{CaO}=0.25



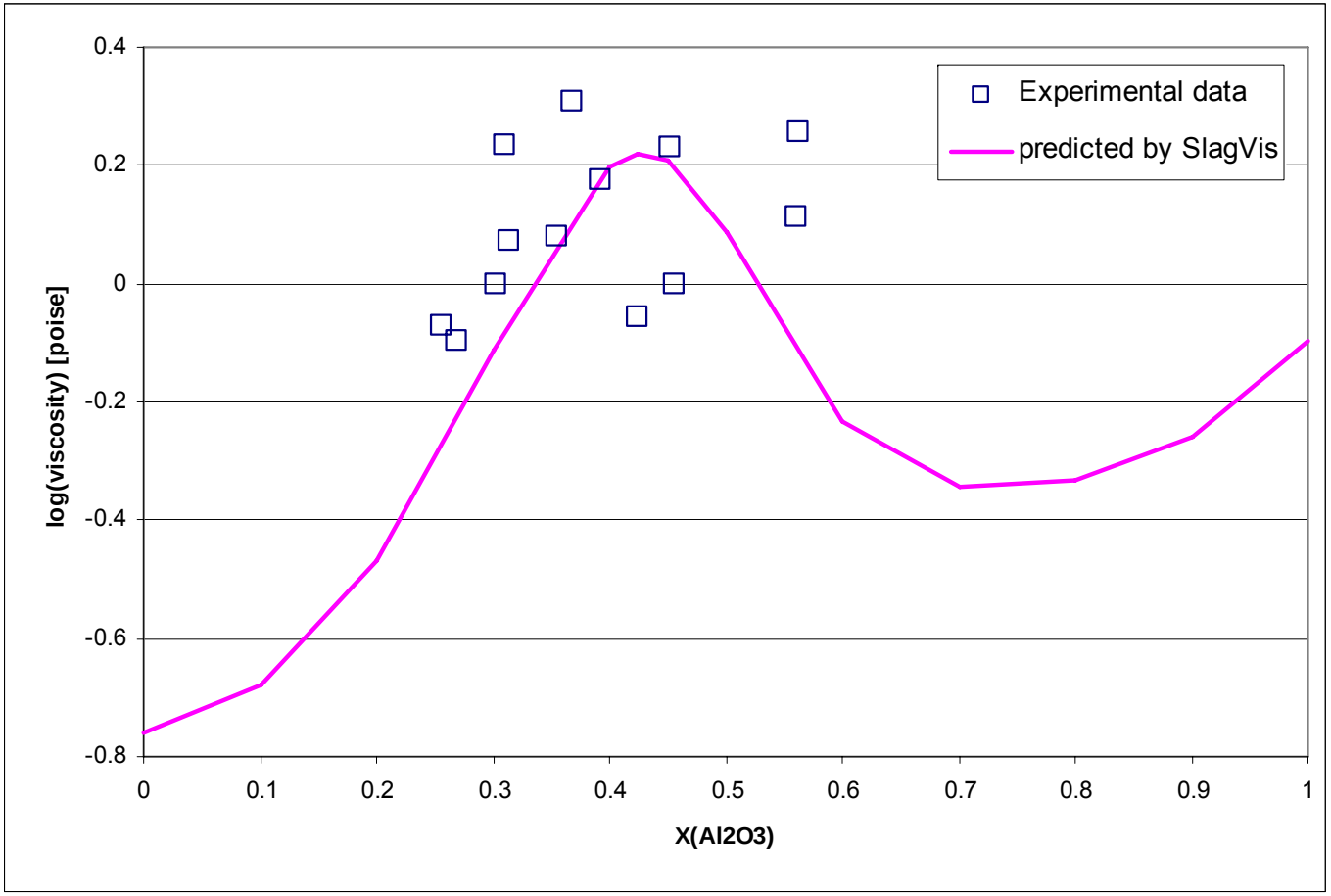
I.3. Effect of composition

Al_2O_3-Na_2O-SiO_2, X_{SiO2}=0.75, 1400°C



I.3. Effect of composition

Al_2O_3-CaO, 1700°C



I.4. Summary

- Neural network approach is essentially suitable for calculating melt viscosity
- Neural network model can be trained with any data (e.g. owner's data)
- Learning algorithm has to be independent of data sorting
- Interpolative capability is good
- Extrapolative capability is reasonable
- Further tests (multi-component melts) are necessary
- Neural network approach can be used in CFD calculations

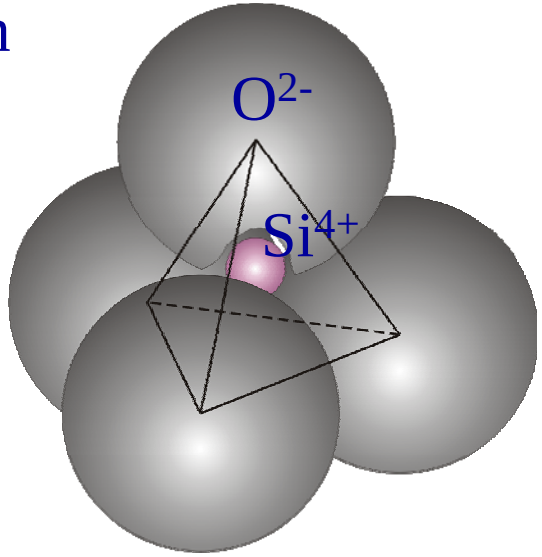
II. Viscosity model

II.0. Introduction

- Viscosity is non-static but dynamic property: “reaction” to applied shear stress
- Theoretically – transport coefficient: transport of momentum
- Viscosity is structure-related property

II.1. Structure of oxide melts

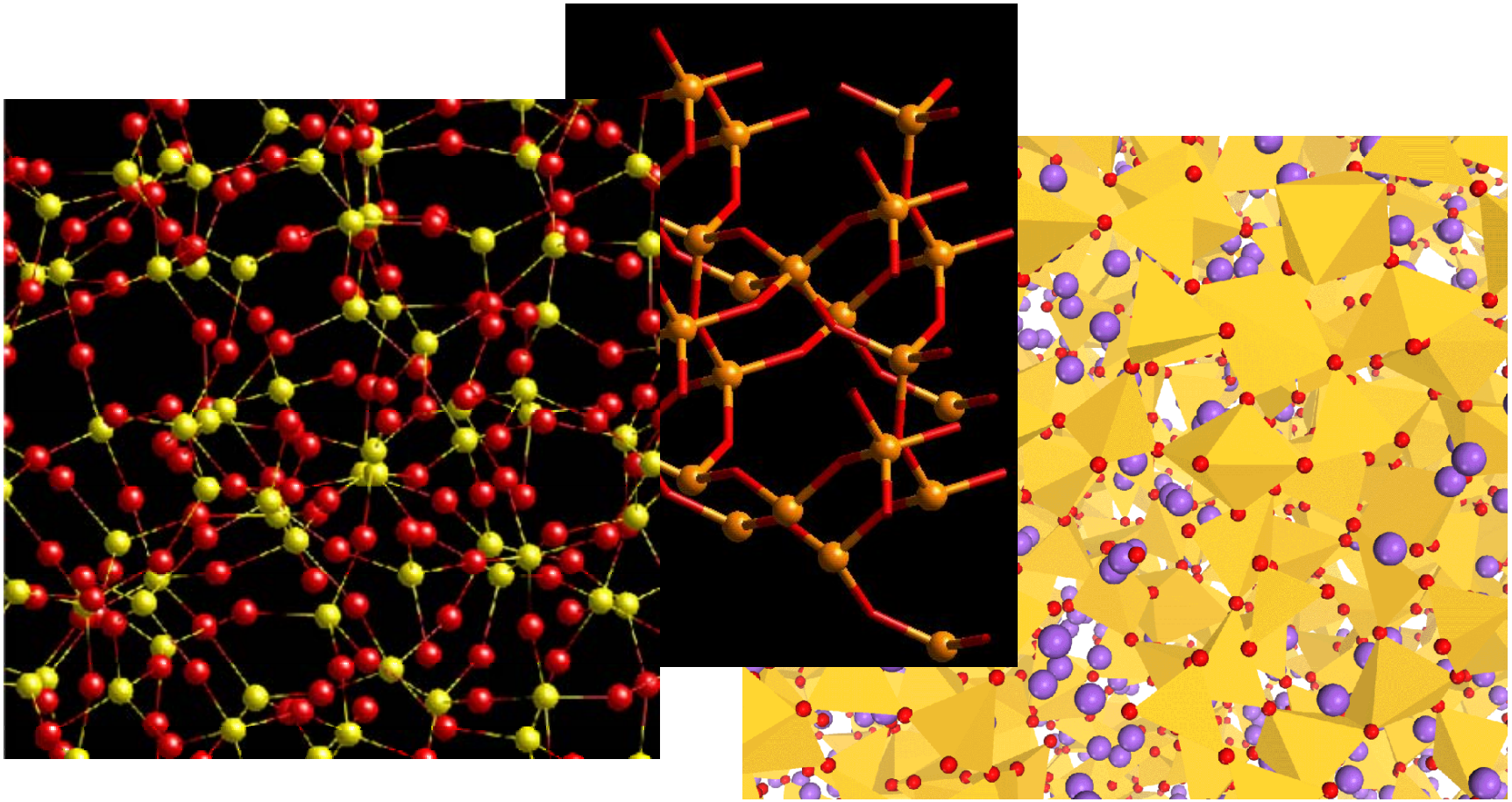
SiO_4^{4-}
tetrahedron



r_{Si-O} = 1.62 Å

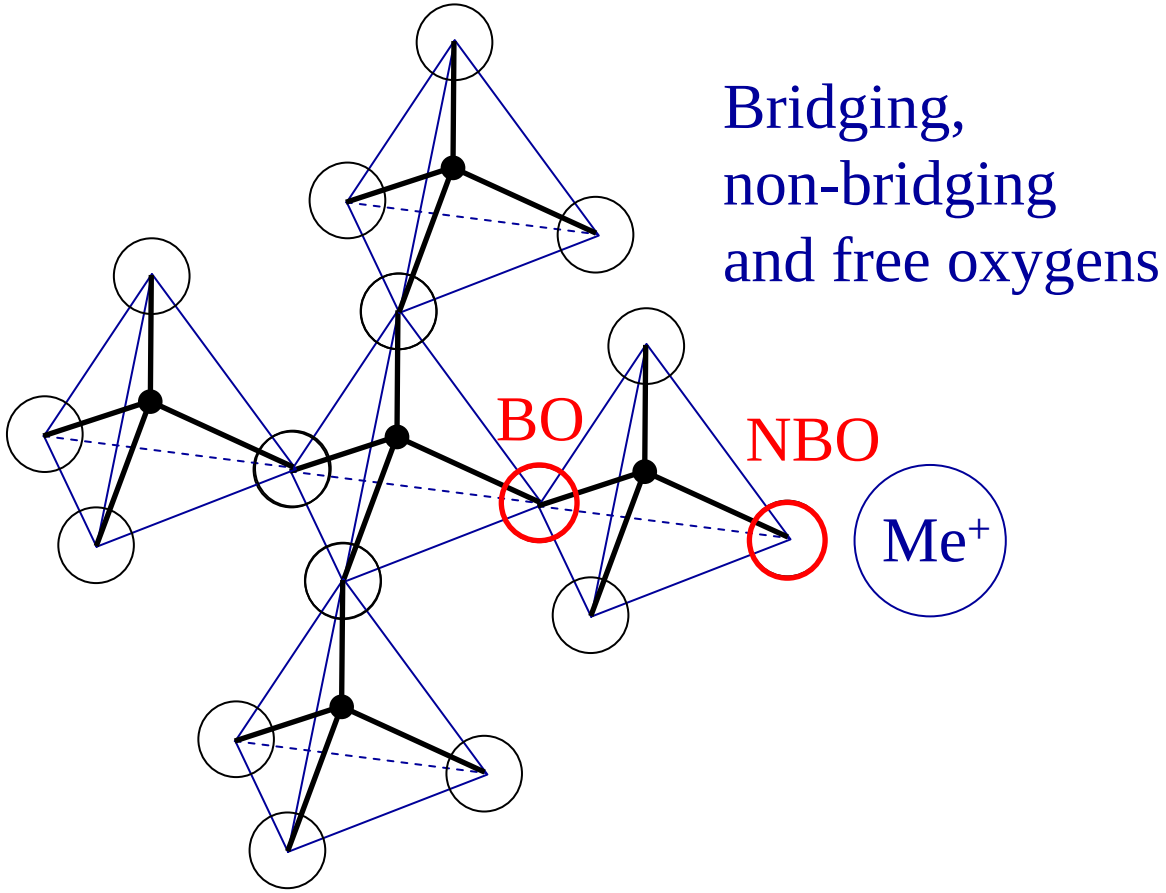
r_{O-O} = 2.65 Å

II.1. Structure of oxide melts



First principles simulation results, Google.com

II.1. Structure of oxide melts



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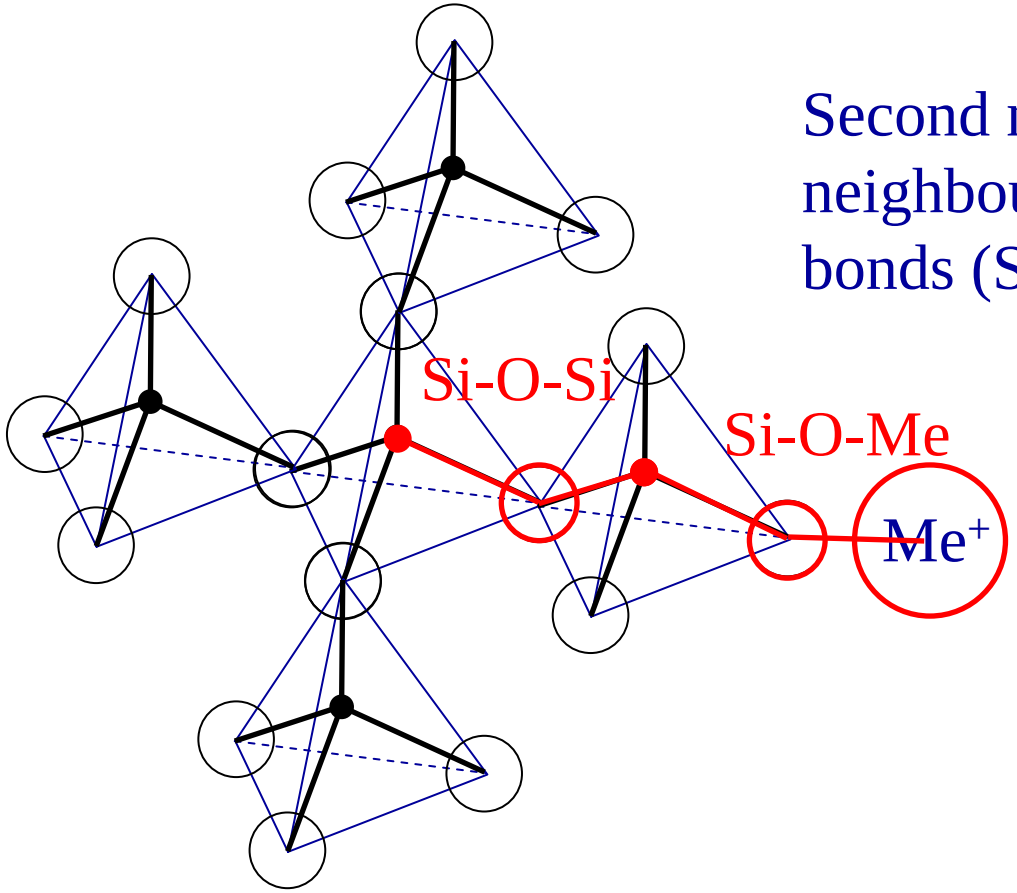

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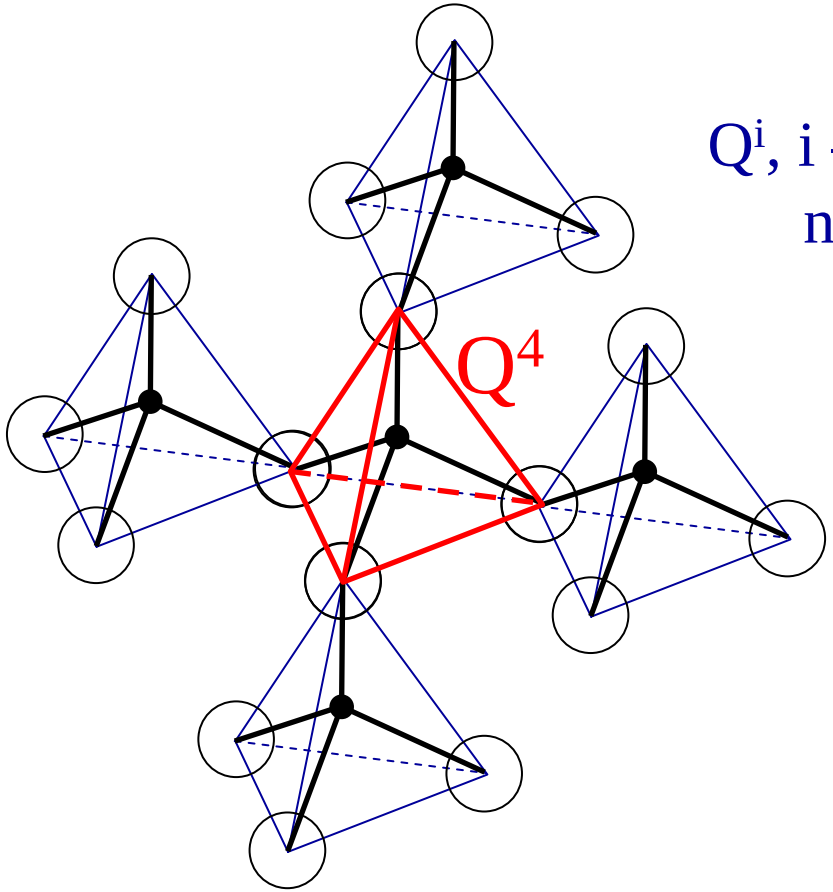
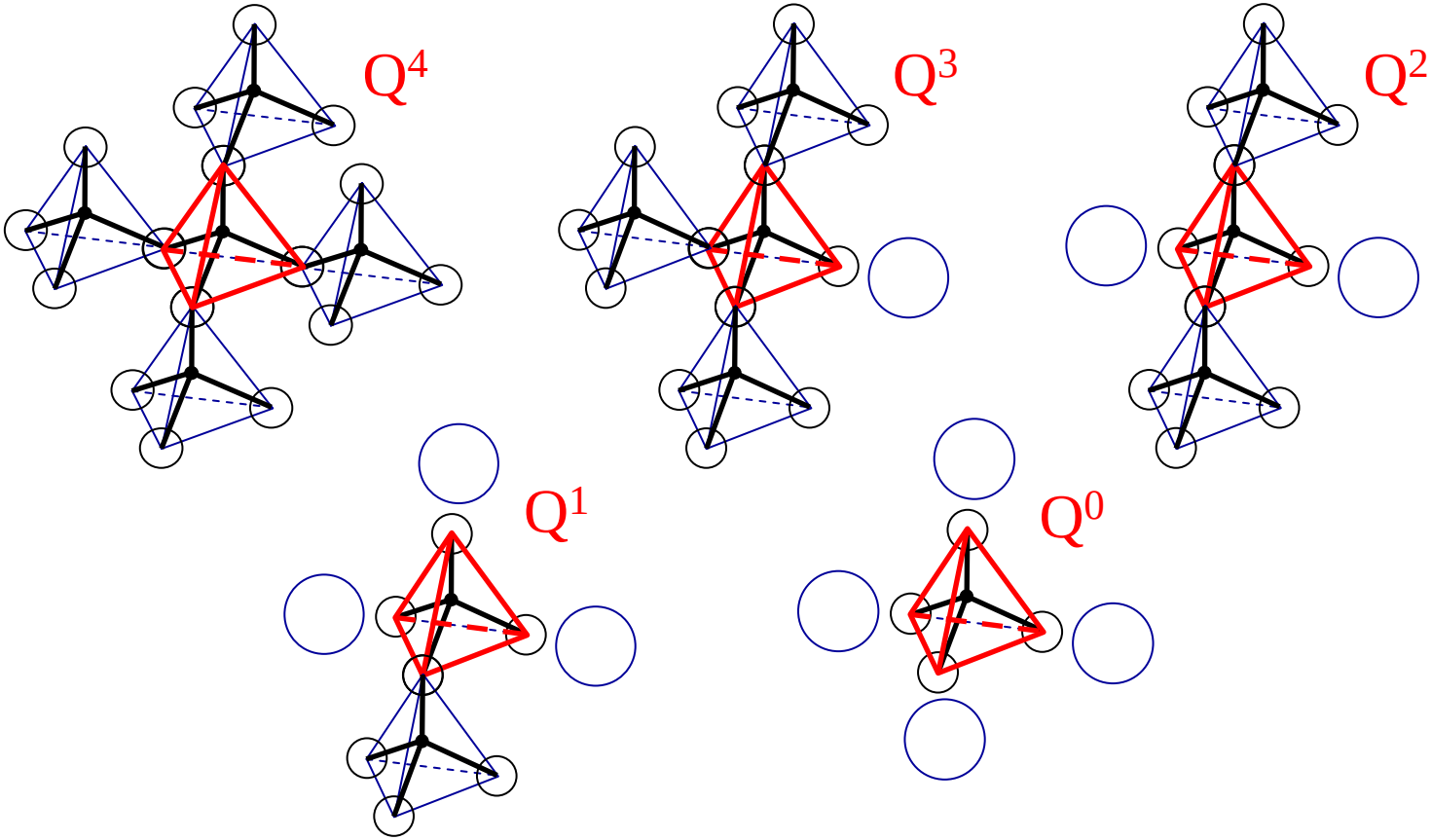


Q^i, i – number of neighbouring tetrahedra

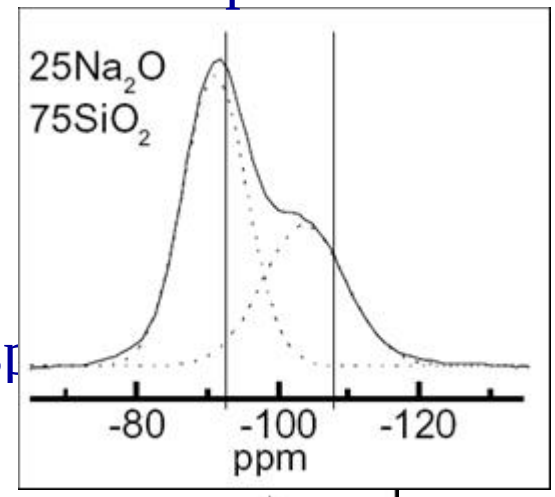
II.1. Structure of oxide melts

Q-species



II.1. Structure of oxide melts

NMR spectra



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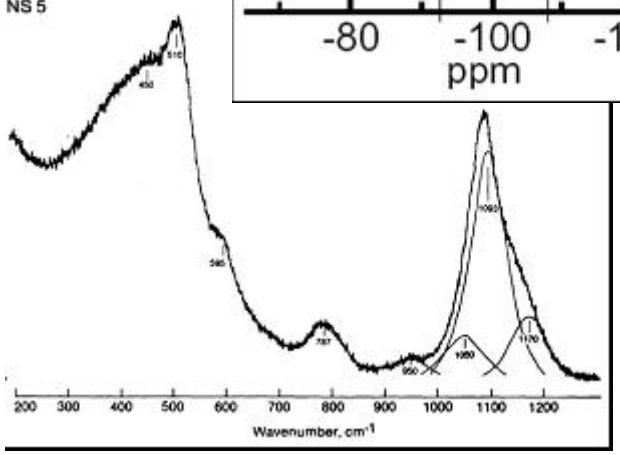
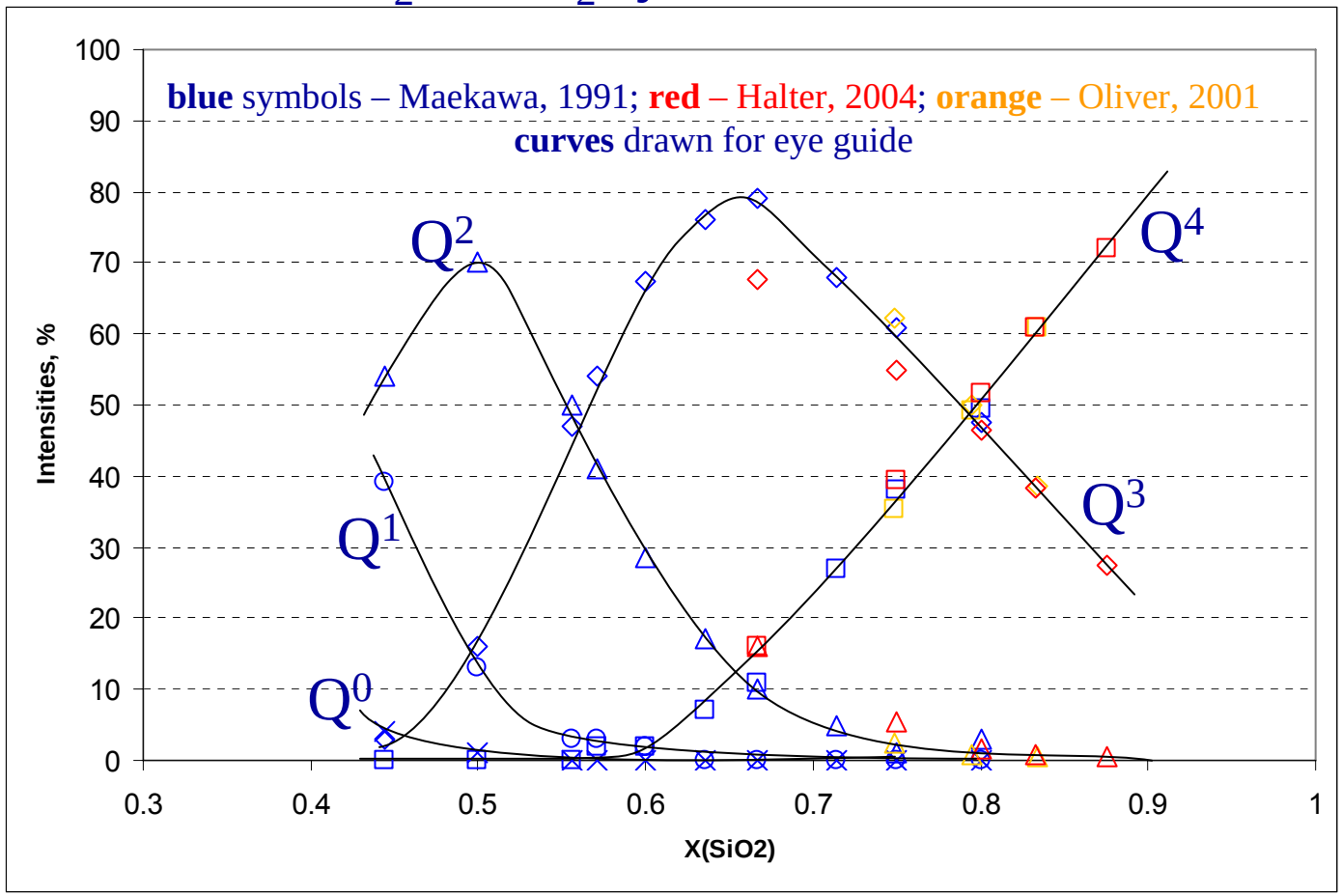


Concentrations of Q^i species as functions of composition & (temperature)

II.1. Structure of oxide melts

Na_2O-SiO_2 system, ~1200°C



II.2. Associate species t/d model

- Yazhenskikh, Hack, Mueller, 2006-2008
- Thermodynamic assessment of Al_2O_3-K_2O-Na_2O-SiO_2 system
- Liquid slag is mixture of associate species: e.g. Na_4SiO_4, K_2SiO_3, $NaAlO_2$ etc.
- Associate species can provide reasonable approximation of melt structure

II.2. Associate species t/d model

- Associate species ↔ Q-species (not always possible; e.g. for QC t/d model)
- For example, in Na_2O-SiO_2 system:

  $Na_4SiO_4 \leftrightarrow Q^0$;

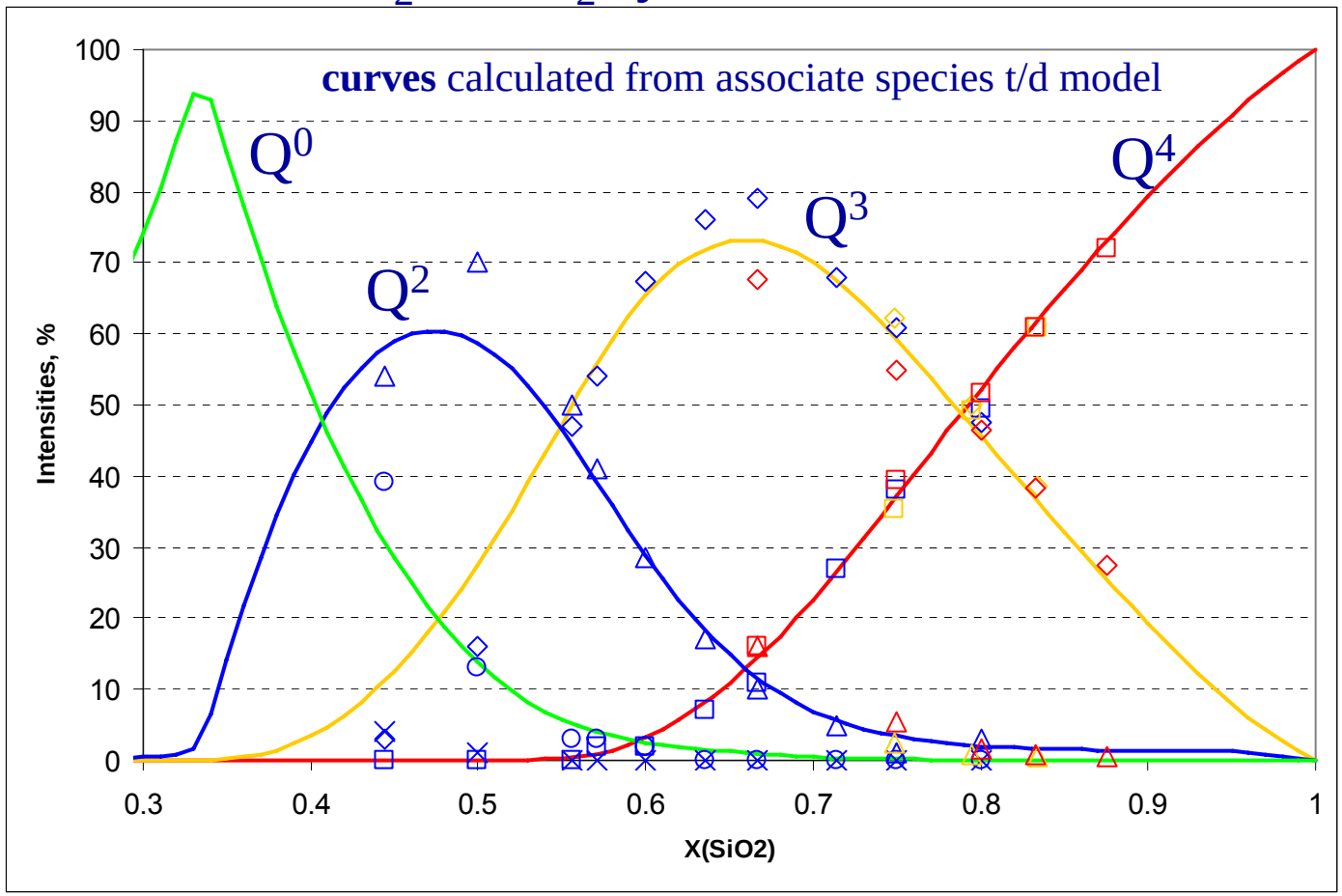
  $Na_2SiO_3 \leftrightarrow Q^2$;

  $Na_2Si_2O_5 \leftrightarrow Q^3$;

  $Si_2O_4 \leftrightarrow Q^4$

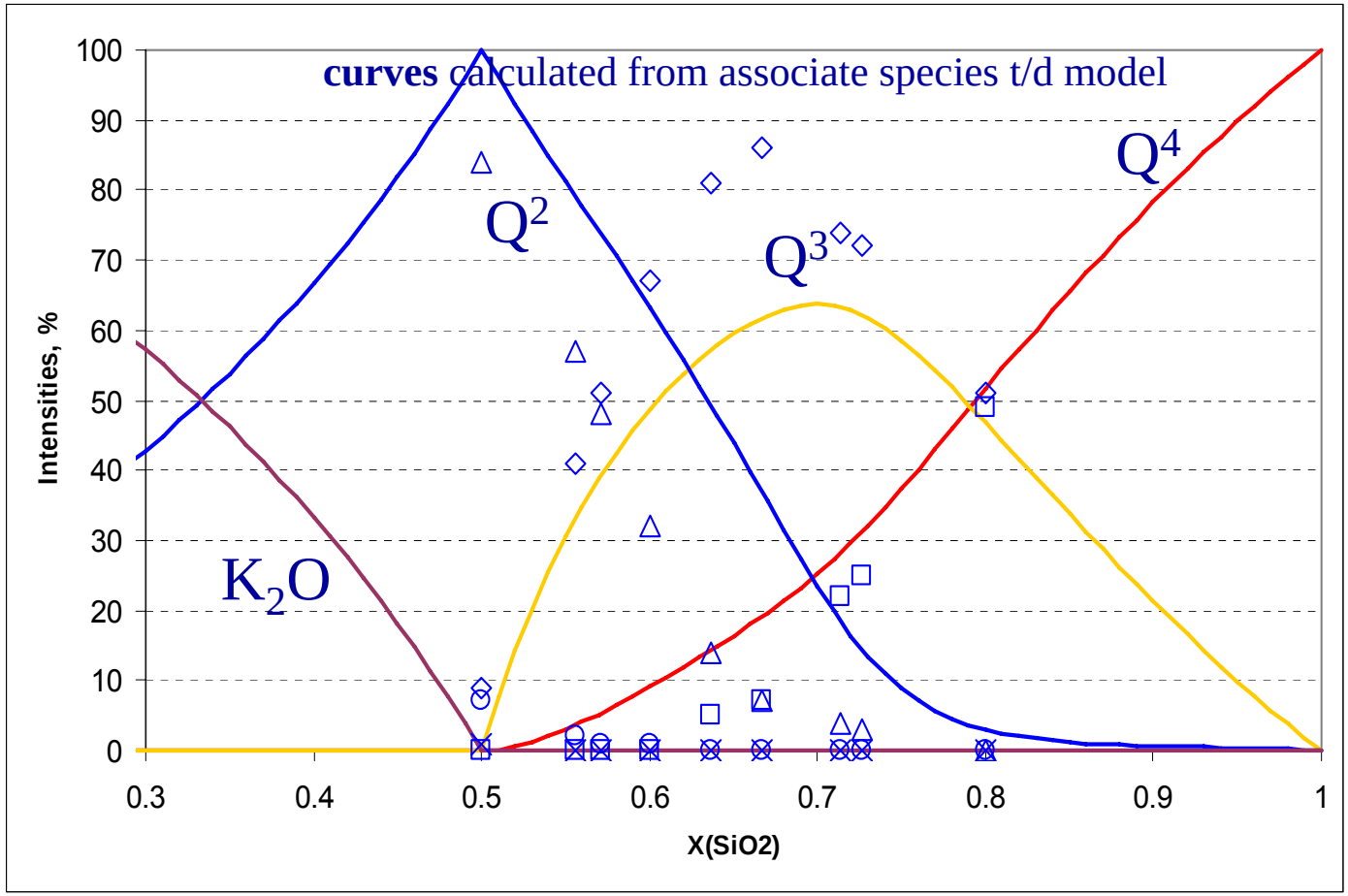
II.2. Associate species t/d model

Na_2O-SiO_2 system, 1200°C



II.2. Associate species t/d model

K_2O-SiO_2 system, 1200°C



II.3. Avramov equation

concentrations of structural units (SU)

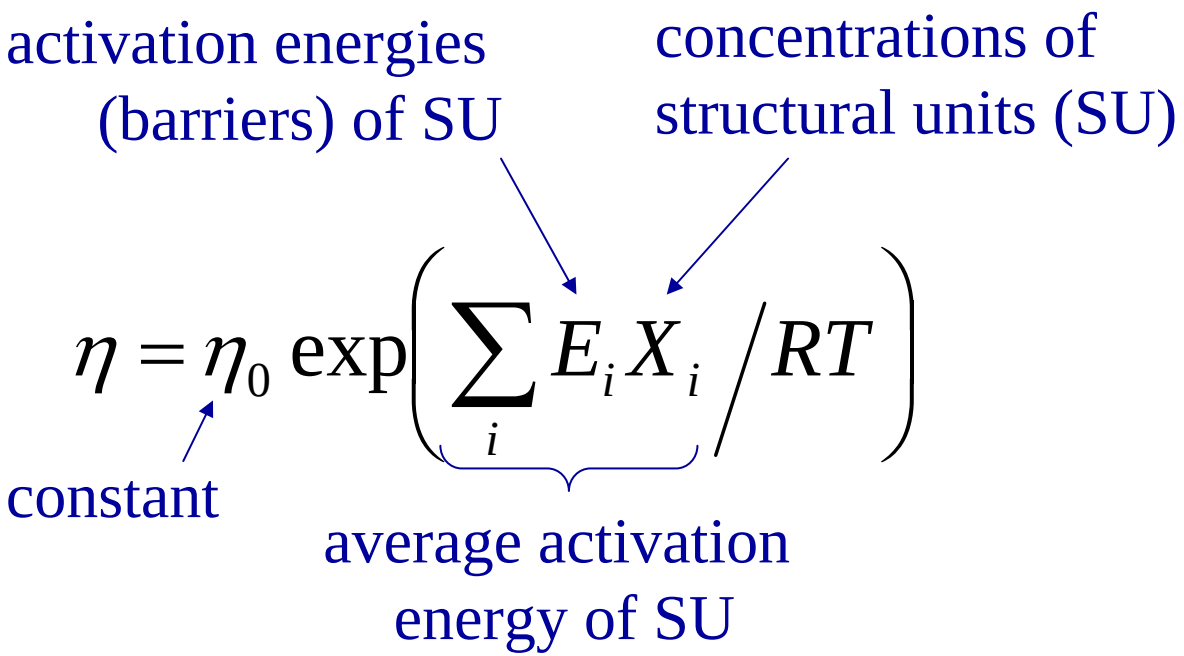
constant

activation energies (barriers) of SU

$$\eta = \frac{\eta_0}{\sum_i X_i \exp\left(-E_i / RT\right)}$$

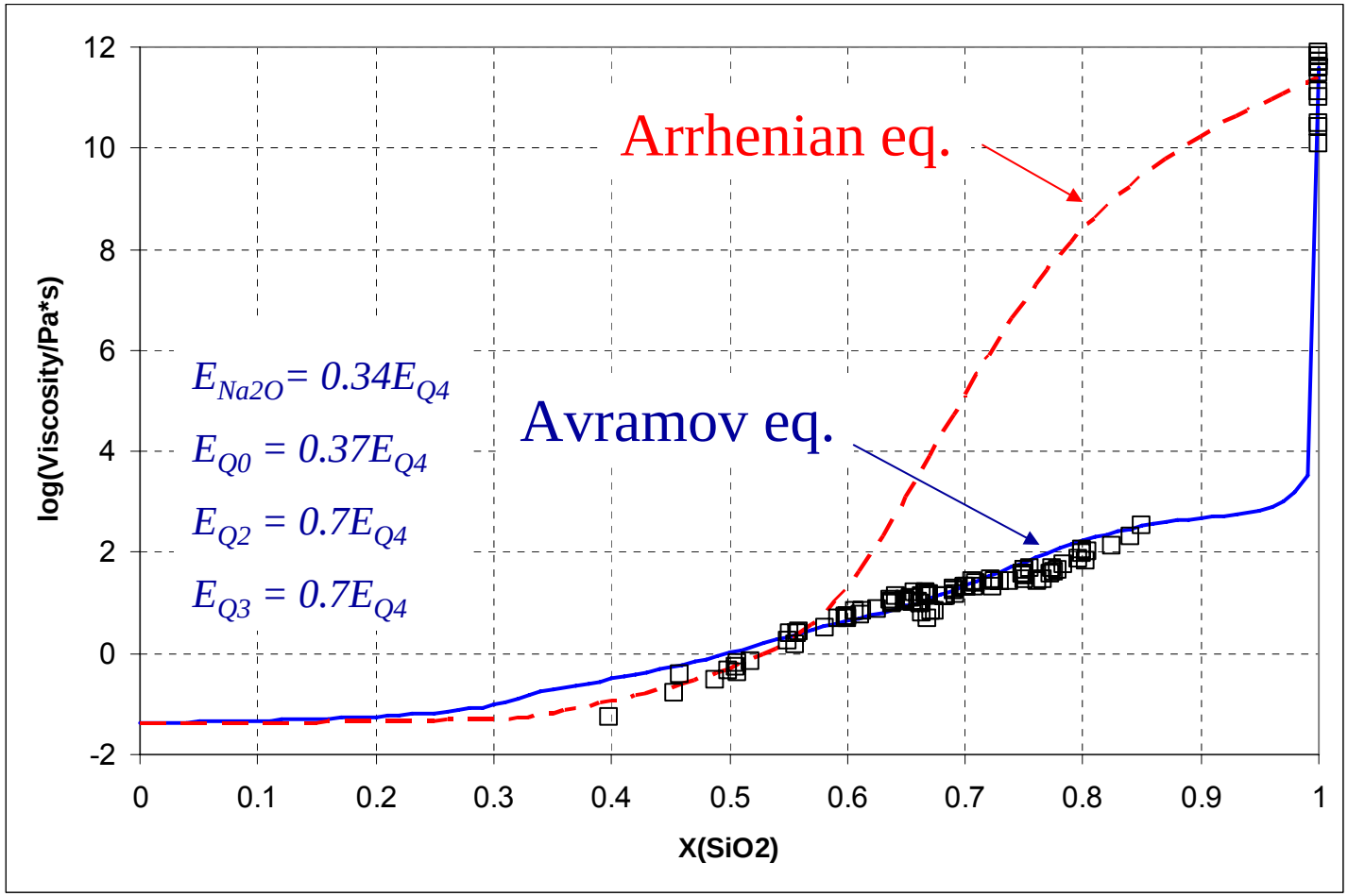
average jump frequency of SU

II.3. Avramov eq vs Arrhenian eq



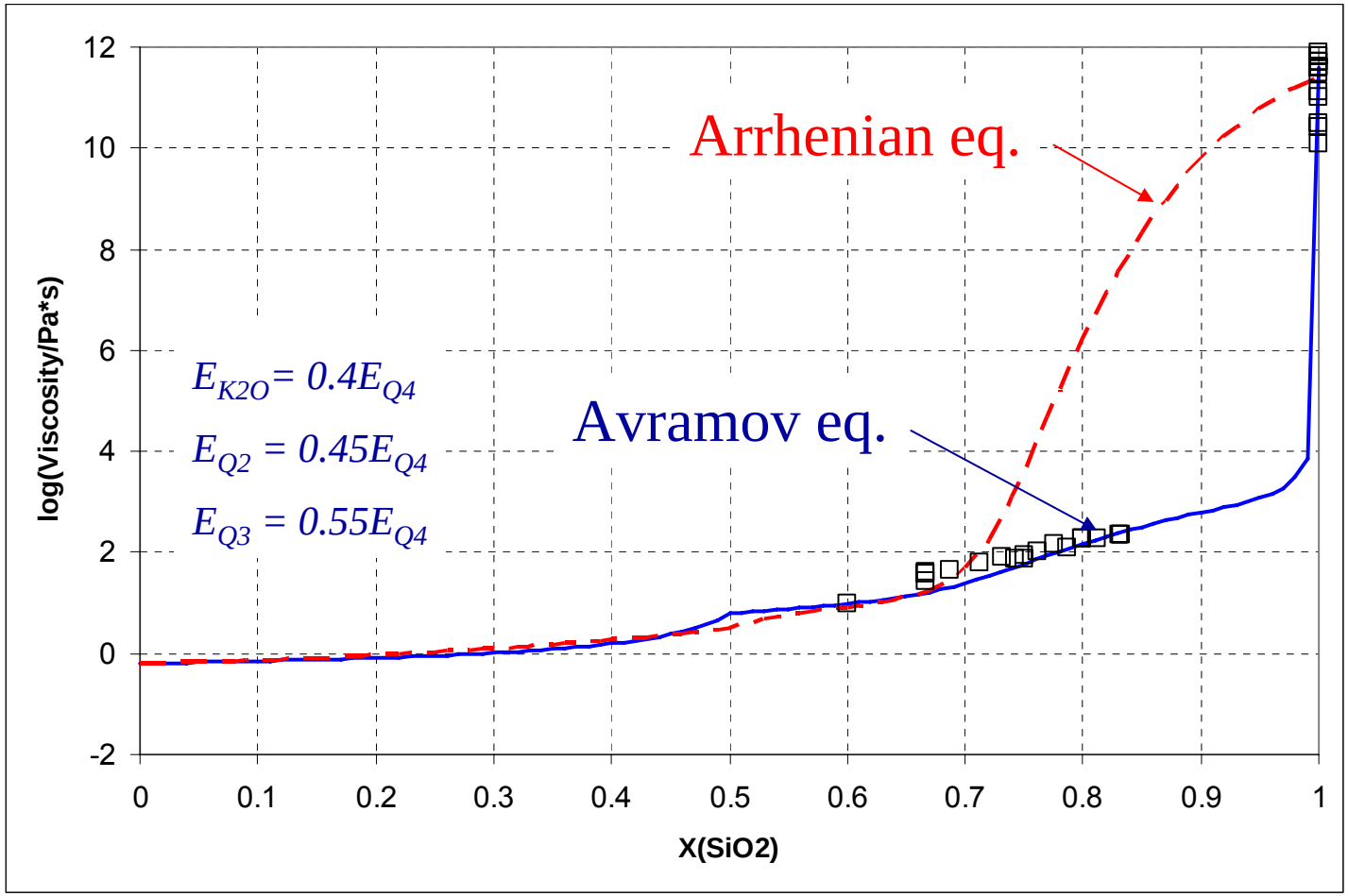
II.4. Viscosities of binary systems

Na_2O-SiO_2 system, 1200°C



II.4. Viscosities of binary systems

K_2O-SiO_2 system, 1200°C



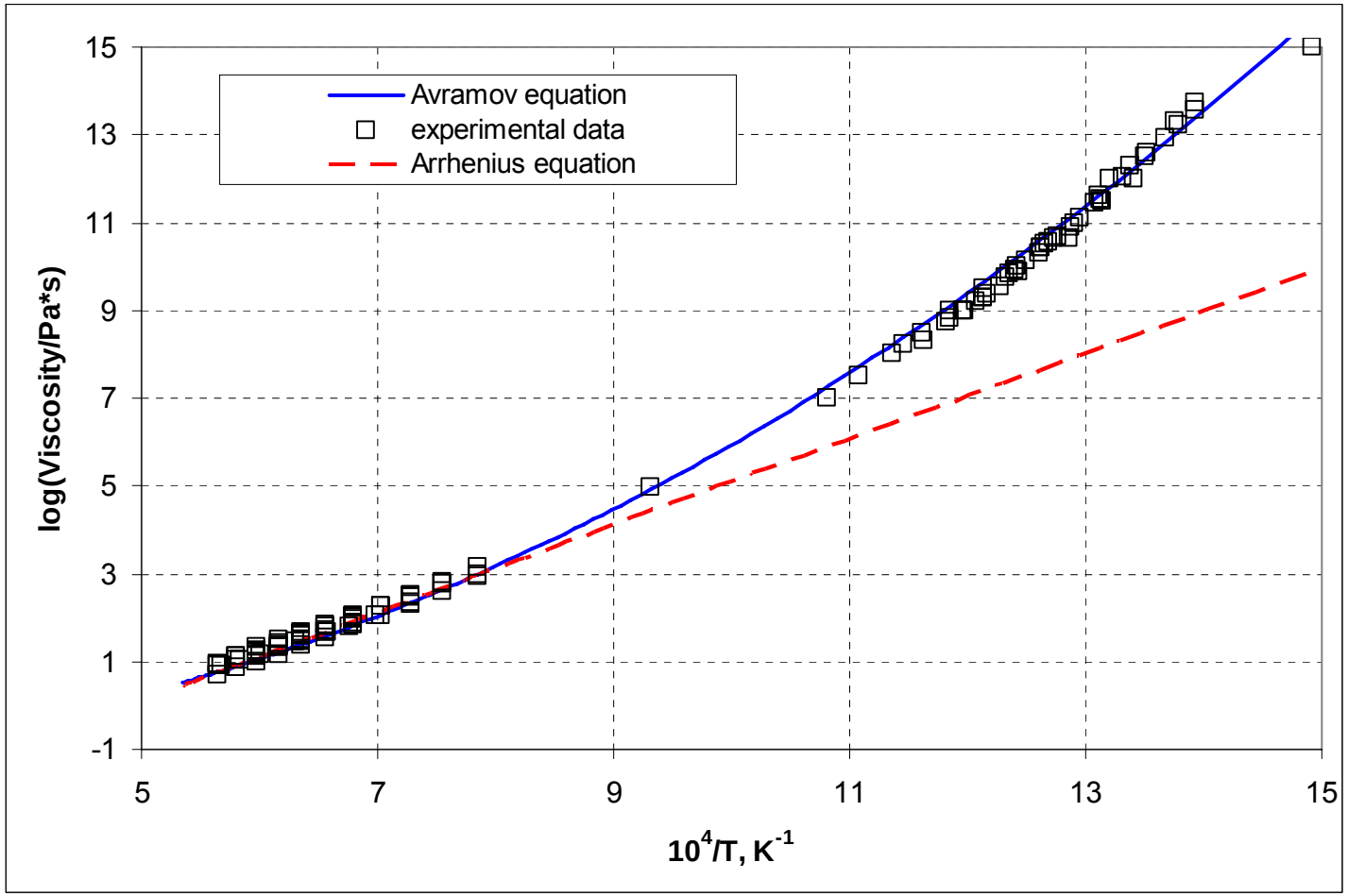
II.5. Temperature dependence

Avramov, JCP, 1991

- Activation energy is distributed (approx. Poisson or Gauss)
- Distribution variance depends on entropy, which in turn depends on temperature
- At low temperatures melt viscosity deviates from linear “logη vs T” dependence

II.5. Temperature dependence

Na_2O-SiO_2 system, X_{SiO2}=0.8



II.6. Summary

- Viscosity model based on Avramov eq can provide better description than Arrhenian-like model
- Melt thermodynamics and viscosity can be described with one set of associate species
- Associate species are to be corrected according to spectroscopic data or/and viscosity data

III. Modelling heterogeneous mixtures

Factors influencing flow behaviour of heterogeneous silicate melts

- Volume fraction of crystals
- Viscosity of liquid phase
- Shape of crystals
- Size of crystals

$$\eta = f\left(\Phi, \eta_L, shape, d_{eff}\right)$$

Roscoe-Einstein equation

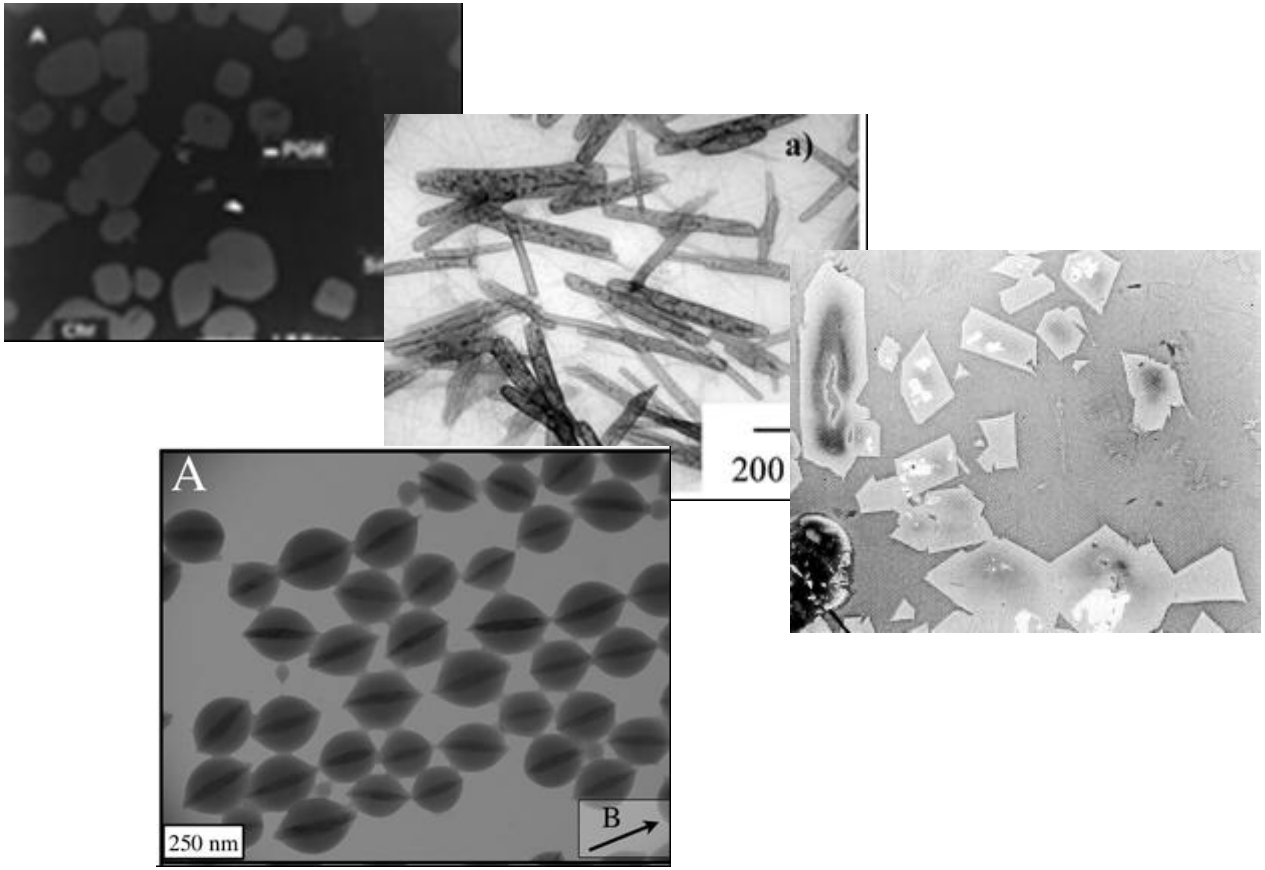
viscosity of remaining liquid

volume fraction of solids

$$\eta = \eta_L \left(1 - R\Phi\right)^{-n}$$

model parameters

Shapes of crystals in silicate melts



Thank you for your attention!