Viscosity of liquid Na-K alloys from molecular dynamics simulations

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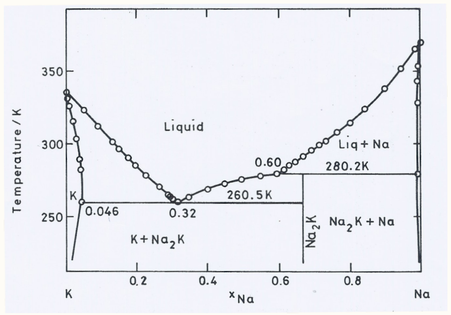
Introduction

The **viscosity** of **liquid metals** and **alloys** is known to be very **low** (lower than water), which leads to several potential applications, like reactor coolant in the case of **Na-K** **[1]**. However, due to **difficulties** to measure such low viscosity, and because alkalis are highly reactive, there are **no recent experimental data** of the viscosity of this alloy. In that respect, **simulation** seems to be an indispensable way to gain access to these data.

The **composition and temperature dependance** of the **shear viscosity** of **Na-K alloys** is analysed

- Nine compositions at 373 K:
  - Na_{10}-K_{90}
  - Na_{20}-K_{80}
  - Na_{30}-K_{70}
  - Na_{40}-K_{60}
  - Na_{50}-K_{50}
  - Na_{60}-K_{40}
  - Na_{70}-K_{30}
  - Na_{80}-K_{20}
  - Na_{90}-K_{10}
- Three different compositions from 280 K to 1000 K:
  - Na_{10}-K_{90}
  - Na_{50}-K_{50}
  - Na_{90}-K_{10}



Phase diagram of the Na-K system **[2]**

Methods

- **Classical molecular dynamics**
- Microcanonical ensemble (N, V, E)
- $N = 2048$ particles
- $\Delta t \approx 0.1$ fs
- 6 000 000 time steps
- Duration of simulation ≈ 1 ns
- **Verlet algorithm**
- **Fiolhais potential** **[3]**

Calculation of the shear viscosity η

1. *Atomic positions $\vec{r}_{ij}$, velocities $\vec{v}_i$ and forces $\vec{f}_{ij}$ are computed*
2. *Off-diagonal stress tensor elements*

$$\downarrow$$

$$\sigma_{\alpha\beta}(t) = -\frac{1}{V}\left(\sum_{i=1}^{N} m_i \vec{v}_{i\alpha}\vec{v}_{i\beta} + \sum_{i=1}^{N-1}\sum_{j>1}^{N} \vec{r}_{ij_\alpha}\vec{f}_{ij_\beta}\right)$$

3. *Autocorrelation* $\rightarrow \langle \sigma_{\alpha\beta}(t_0)\cdot\sigma_{\alpha\beta}(t_0+t)\rangle$
4. ***Green-Kubo relation***

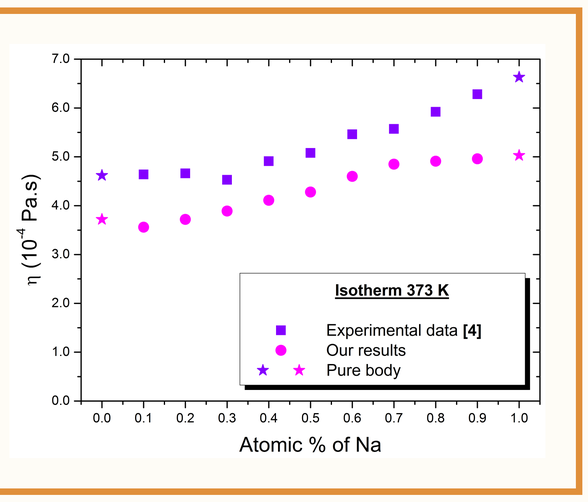
$$\Downarrow$$

$$\eta_{\alpha\beta} = \frac{V}{k_B T}\int_0^\infty \langle \sigma_{\alpha\beta}(t_0)\sigma_{\alpha\beta}(t_0+t)\rangle dt$$

To improve the statistics:

- Average over six stress tensor elements: σ_{xy}, σ_{yz}, σ_{xz}, $\frac{1}{2}(\sigma_{xx}-\sigma_{yy})$, $\frac{1}{2}(\sigma_{yy}-\sigma_{zz})$, $\frac{1}{2}(\sigma_{xx}-\sigma_{zz})$
- Successive shifts of time origin in Green-Kubo relation
- Average over eight different runs

Results



Viscosity of Na-K alloy vs concentration at 373 K

Statistical error on the simulation results is about 5 %

Agreement with experiment [4]

$$\Downarrow$$

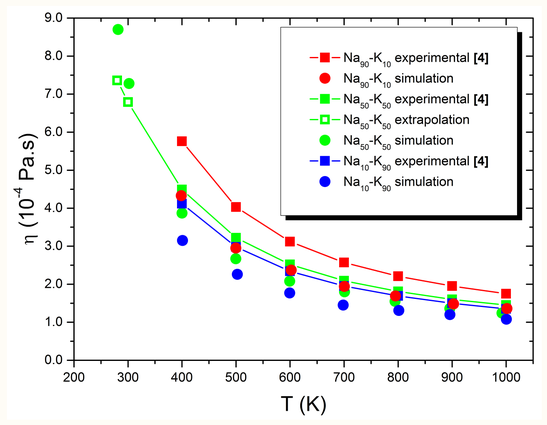
Shift but **same trend**

↪ Inaccuracy of the potential

↪ Impurities in the experiments

$\Rightarrow$ η increases monotonously with Na concentration

$\Leftarrow$
- T ↘ $\Rightarrow$ η ↗
- Na ↘ $\Rightarrow$ η ↘



Viscosity of Na-K alloy vs temperature for three studied compositions

Chemical order:

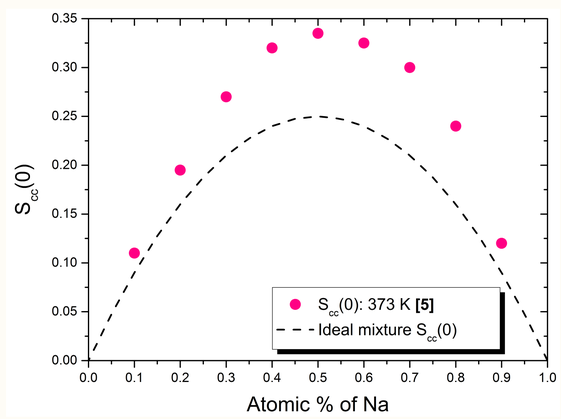
- Bathia-Thornton partial structure factor $S_{cc}(q=0) > c_{Na}c_K$

$$\downarrow$$

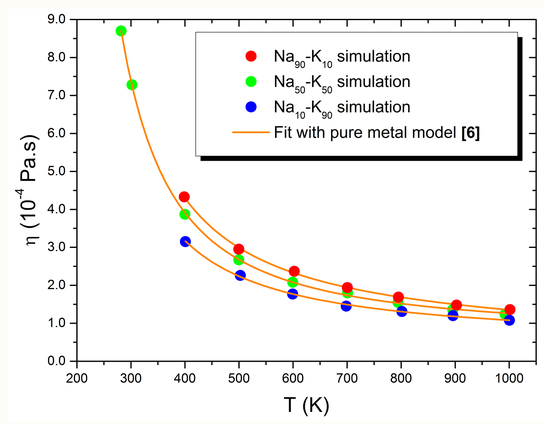
Homocoordination tendencies

- **Weak chemical order** **[5]**

$\Rightarrow$



Values of $S_{cc}(0)$ for Na-K alloy vs composition at 373 K



Simulation data fitted by pure metal model

$\Leftarrow$

Fit by equation for pure metal **[6]**:

$$\eta(T) = \left(AT^2 + C\right)e^{B/T}$$

	A (Pa.s.K^{-2})	B (K)	C (Pa.s)
Na_{90}-K_{10}	-7.94×10^{-12}	706.11	7.00×10^{-5}
Na_{50}-K_{50}	9.03×10^{-13}	762.75	6.00×10^{-5}
Na_{10}-K_{90}	-2.94×10^{-12}	688.75	6.00×10^{-5}

Conclusion

Simulation and experimental data follow the **same trend**. In qualitative terms, the **Fiolhais potential** seems to be **successful** to compute the **shear viscosity** of **Na-K**.

As a **consequence of the weak chemical order** of the alloy:

- The evolution of the **viscosity of Na-K versus concentration** is **rather smooth**.
- The evolution of the **viscosity of Na-K versus temperature** is similar to that of a **pure one component fluid**. The **shear viscosity** can then be calculated by the **relation** proposed by Meyer *et al.* for **liquid Na** **[6]**.

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