

Molten Salt Rheology of FLiNaK from Molecular Dynamics with Uncertainty Quantification

Minki Kim^a, Wonjae Lee^a, Yunbum Park^b, Taewoo Kim^b, Jaekwang Kim^{a*}

^aDepartment of Mechanical and Design Engineering, Hongik University, Sejong, S. Korea

^bKorea Atomic Energy Research Institute, 111, Daedeok-Daero 989 beon-gil, Yuseong-Gu, Daejeon., S. Korea

*Corresponding author: jk12@hongik.ac.kr

***Keywords :** Nuclear Power Plants, Molten FLiNaK, Shear viscosity, Molecular dynamics, Uncertainty quantification

1. Introduction

High-temperature viscosity data for molten FLiNaK are essential for the thermal-hydraulic analysis and component design of molten salt reactor systems. However, reliable experimental measurements remain limited because of the high operating temperatures and the corrosive nature of fluoride salts. As a result, molecular dynamics simulation has become a useful approach for predicting transport properties under reactor-relevant conditions.

In this study, the shear viscosity of molten FLiNaK was evaluated over 750–1000 K using molecular dynamics and the Green–Kubo formalism. A rigid-ion interaction model was adopted to calculate the stress autocorrelation function and its time integral. In addition to predicting the temperature-dependent viscosity of FLiNaK, this study assesses the reliability of the calculated values through uncertainty quantification. Details of the viscosity calculation and uncertainty analysis are provided in the Methods section.

2. Methods

Molecular dynamics simulations of molten FLiNaK were performed using LAMMPS under three-dimensional periodic boundary conditions. The eutectic salt composition was constructed by randomly assigning Li⁺, Na, and K⁺ ions to the cation sites with F⁻ ions occupying the anion sites. Interionic interactions were described using a rigid-ion potential consisting of long-range Coulomb interactions and short-range Buckingham-type terms. The long-range electrostatic contribution was treated with a mesh-based solver. Simulations were carried out at 750 K, 800 K, 850 K, 900 K, 950 K, and 1000 K.

For each temperature, the system was first equilibrated to obtain the corresponding liquid-state structure and density, and the viscosity calculation was then performed from an NVE production run. The integration time step was set to 1 fs. During the production stage, the off-diagonal stress tensor components P_{xy} , P_{xz} , and P_{yz} were recorded. The shear viscosity was calculated from the Green–Kubo relation by integrating the autocorrelation function of the off-diagonal stress components and averaging the three independent contributions.

The total uncertainty was separated into statistical, parametric, and model-form contributions. Statistical uncertainty was estimated from block averaging of independent trajectory segments and from the sensitivity of the Green–Kubo integral to the cutoff time. Parametric uncertainty was evaluated by perturbing selected short-range potential parameters within reasonable bounds and examining the corresponding changes in viscosity. Model-form uncertainty was discussed based on the intrinsic limitation of the rigid-ion approximation, particularly the neglect of polarization effects.

3. Results and Discussion

The predicted shear viscosity of molten FLiNaK decreased monotonically with increasing temperature over the entire range of 750–1000 K. This trend is physically consistent with the enhanced ionic mobility at elevated temperatures. At lower temperatures, the stress autocorrelation function decayed more slowly, resulting in a larger Green–Kubo integral and thus a higher viscosity. At higher temperatures, faster stress relaxation led to smaller viscosity values.

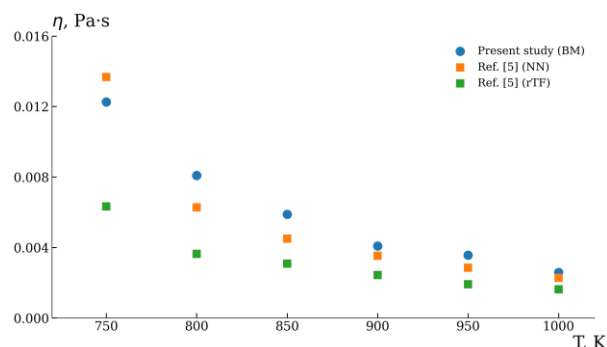


Figure 1 Temperature-dependent shear viscosity of molten FLiNaK predicted in the present study using the BM model, compared with NN and rTF results reported in Ref. [5].

Figure 1 compares the BM-based viscosity predictions obtained in the present study with the NN and rTF results reported in Ref. [5]. The present results show the expected monotonic decrease in shear viscosity with increasing temperature and follow the overall temperature dependence reported in the literature. Over the investigated range, the predicted values remain

within the same order of magnitude as the reference results, while some quantitative differences are still observed. These discrepancies are attributed to differences in interaction models, simulation settings, and finite sampling effects associated with the Green–Kubo calculation. In particular, the deviations at intermediate and lower temperatures are considered to be related to slower stress autocorrelation decay and the increased sensitivity of the viscosity integral to the selected cutoff time.

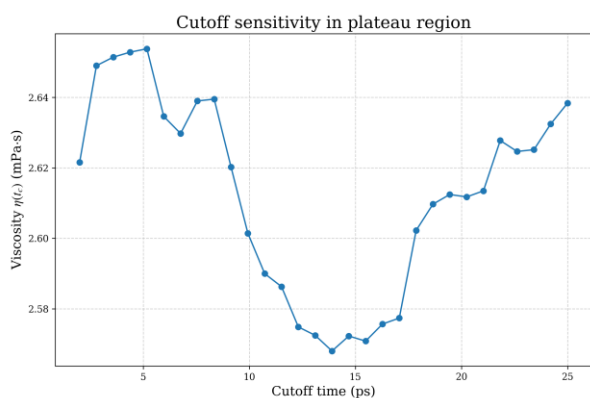


Figure 2 Cutoff-time sensitivity of the Green–Kubo viscosity in the plateau region for molten FLiNaK at 1000 K.

The uncertainty analysis indicates that the reliability of the predicted viscosity depends on both temperature and sampling conditions. Statistical uncertainty becomes more pronounced at lower temperatures because the slow decay of the stress autocorrelation function makes the Green–Kubo integral more sensitive to trajectory length and cutoff-time selection. Parametric uncertainty reflects the dependence of the calculated viscosity on the short-range potential parameters, whereas model-form uncertainty represents the systematic limitation of the rigid-ion description. Figure 2 illustrates the sensitivity of the predicted viscosity to the selected cutoff time within the plateau region at 1000 K, showing that the final viscosity remains within a relatively narrow range over the selected cutoff window. This result supports the stability of the plateau-based viscosity estimate and provides a practical measure of the statistical uncertainty associated with the Green–Kubo integration.

4. Conclusion

This study evaluated the shear viscosity of molten FLiNaK over 750–1000 K using molecular dynamics and the Green–Kubo formalism. The calculated viscosity showed the expected monotonic decrease with increasing temperature. In addition to the viscosity prediction itself, the present work examined uncertainty by separating statistical, parametric, and model-form contributions. The results suggest that both finite sampling and model assumptions can noticeably affect the predicted viscosity,

especially at lower temperatures. The present workflow provides a practical basis for reactor-relevant molten salt property evaluation and may be further improved through longer simulations and higher-fidelity interaction models.

REFERENCES

- [1] D. O. Zakiryanov, *Applying the Born-Mayer model to describe the physicochemical properties of FLiNaK ternary melt*, Computational and Theoretical Chemistry, Vol. 1219, Article 113951, 2023.
- [2] L. Langford, N. Winner, A. Hwang, H. Williams, L. Vergari, R.O. Scarlet, and M. Asta, Constant-potential Molecular Dynamics Simulations of Molten Salt Double Layers for FLiBe and FLiNaK, *J. Chem. Phys.*, Vol. 157, Art. 094705, 2022, doi:10.1063/5.0097697.
- [3] J. Lu, S. Yang, G. Pan, J. Ding, S. Liu, and W. Wang, Thermal and Transport Properties of Molten Chloride Salts with Polarization Effect on Microstructure, *Energies*, Vol. 14, Art. 746, 2021.
- [4] N. Rybin, D. Maksimov, Y. Zaikov, and A. Shapeev, Thermophysical Properties of Molten FLiNaK: A Moment Tensor Potential Approach, *Journal of Molecular Liquids*, Vol. 410, Art. 125402, 2024.
- [5] S.-C. Lee, Y. Zhai, Z. Li, N.P. Walter, M. Rose, B.J. Heuser, and Y. Zhang, Comparative Studies of the Structural and Transport Properties of Molten Salt FLiNaK Using the Machine-Learned Neural Network and Reparametrized Classical Forcefields, *Journal of Physical Chemistry B*, Vol. 125, No. 37, pp. 10562–10570, 2021.