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Research Article

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Posted Date: July 6th, 2026

DOI: <https://doi.org/10.21203/rs.3.rs-10223172/v1>

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Additional Declarations: No competing interests reported.

Version of Record: A version of this preprint was published at International Journal of Thermophysics on October 12th, 2026. See the published version at <https://doi.org/10.1007/s10765-026-03842-3>.

Integration of a Structural Coherence Model with a Semi-Universal Machine Learning Interatomic Potential to Predict Molten Salt Thermal Conductivity

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Abstract

The accurate design and performance assessment of energy generation systems that use molten salts as working fluids require accurate characterization of thermal conductivity, with a sufficiently low uncertainty. However, experimental data on several molten salt systems, of interest for various energy generation applications, are generally lacking. Physics-informed models, as well as molecular dynamic (MD) simulations, have the potential to populate gaps in current thermophysical property databases. Existing predictive models using MD simulation (Green-Kubo, non-equilibrium MD) take significantly longer to predict thermal conductivity than classical MD models that otherwise predict salt density with reasonable accuracy. We present an approach that uses the SuperSalt machine-learning interatomic potentials (MLIPs) for chloride salts, as well as a structural coherence model (SCM) to predict thermal conductivity of molten salts using volumetric heat capacity, adiabatic speed of sound, and the partial pair distribution function. The SCM uses this function to predict the mean free path of thermal transport and applies generalized kinetic theory to predict thermal conductivity near the melting temperature, achieving results 1–2 orders of magnitude faster

than existing MD methods. By also predicting the isothermal bulk modulus, the temperature dependence of thermal conductivity above the melting temperature can be determined. This integrated SCM-MLIP (ISM) methodology serves as a proof of concept for using MLIPs to predict thermal properties within current experimental uncertainties at a fraction of the computational cost. This enables investigation into thermal conductivity as well as other thermophysical properties across a variety of molten salt compositions. When coupled with targeted experimental studies, the ISM methodology can help contribute to existing databases necessary to safely implement molten salts into power generation systems.

Keywords: Thermal transport, molecular dynamics, machine learning interatomic potentials, molten chloride salts

1 Introduction

To support the accurate design and safe operation of energy generation systems utilizing molten salt, characterization of the working fluid with sufficiently low uncertainty is required. While low-melting-point nitrate salts, such as those used in concentrated solar power systems, are well characterized and have been used on a commercial scale [88], measurements of halide salts are challenged by higher melting temperatures and a wide range of compositional variation. In the case of molten salt reactors (MSRs)—especially those using actinide-bearing salts where thermophysical data is generally more limited [89]—accurate prediction of thermophysical properties such as specific heat capacity (c_p), density (ρ), viscosity (μ), and thermal conductivity (k) is needed [66]. Thermal conductivity and heat capacity are especially important for predicting MSR transient responses and heat exchanger behavior [63]. In the case of MSRs, understanding the change in thermal conductivity as the composition changes with fuel burnup and fission product accumulation is essential to safely account for changes in reactor operating parameters over its lifetime [13, 59, 63]. However, current experimental methods struggle to provide consistent correlations and trends compared to models (Figure 1) due to high temperatures, the highly corrosive nature of molten salts, and the difficulty associated with isolating conduction as the variable being measured among convective and radiative heat transfer modes. [55, 58, 81].

There is a marked scarcity of thermal conductivity data currently available. Among the current 976 salt compositions listed by the Molten Salt Database - Thermophysical v4.0 (MSD-TP), only 32 compositions have thermal conductivity measurements recorded, with only 9 of the 32 being actinide-bearing mixtures [8]. Of these values, an average of 18% uncertainty is cited. The spread of measured and predicted thermal conductivity is illustrated in Fig. 1 for the case of LiF-NaF-KF (0.462-0.115-0.42 mol%), also known as FLiNaK, in which concerted multi-group efforts continue to yield notable variation [55]. High uncertainty is also typical across chloride salts [9]. This disagreement between measurement and modeling approaches and results is compounded by the lack of an established theory of liquid thermal conductivity to interpret results, emphasizing the need for improved resolution of microscale energy transport

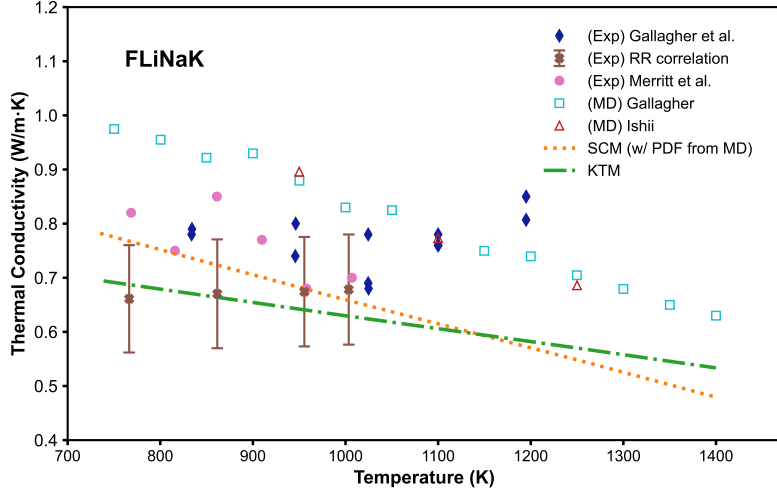


Fig. 1: Comparison of predicted FLiNaK (0.462-0.115-0.42 mol % LiF-NaF-KF) thermal conductivity compared to experimental measurements [29, 55, 62], MD simulations [40], and predictive models (SCM–structural coherence model [67], KTM–kinetic theory model [30]).

mechanisms in molten salts [67]. The scarcity and inaccuracy of thermal conductivity pose a risk to bottleneck MSR development, parameterization, and optimization [46].

1.1 Molecular Dynamic (MD) Models

Molecular dynamics (MD) simulations provide an alternative approach to fill the gap in thermal conductivity data in databases [68]. However, existing particle-tracking models like the Green-Kubo equilibrium MD (EMD) method as well as both standard and reverse non-equilibrium MD (NEMD and RNEMD) often mispredict thermal conductivity [15, 32, 99] despite using highly computationally intensive *ab initio* MD (AIMD) to improve accuracy [60]. The EMD method can be executed in a small simulation space, though it requires long time scales, while the opposite is true for NEMD methods [25]. While NEMD methods previously required simulation sizes that were too large for AIMD, the advent of Machine Learning Interatomic Potentials (MLIP), which are large neural networks trained on DFT data, created an alternative lower computational-cost pathway to AIMD while maintaining similar accuracy. This allows for NEMD methods to be performed at the size scale needed to avoid finite-size effects [10] with high enough accuracy to capture behavior of salts that cannot be simulated accurately using traditional classical potentials such as Born-Mayer-Huggins-Tosi-Fumi (BMHTF). However, the incorporation of actinide-bearing compounds computed using density functional theory (DFT) to create training data is difficult due to their 5f orbitals and thus highly complex electronic structure [48, 65], and calculation of thermal conductivity via EMD, NEMD, or RNEMD using an MLIP can still be computationally expensive when studying a complex salt with a large

number of cation species expected as uranium fission products [47, 77, 92]. MD predictive methods have produced data with comparable accuracy to experimental data, but their computational cost—even reduced with MLIP usage—remains a barrier in using them to fill gaps in the MSD-TP.

1.2 Kinetic Theory and Structural Coherence Models (KTM and SCM)

In addition to direct MD simulations of atomistic energy transport, theoretical descriptions of thermal conductivity (k) in liquids attempt to encode the underlying mechanisms in a physics-based analytical model. A dominant framework for thermal conductivity models is the consideration of energy transport by characterizing the energy carriers. This involves defining the effective attributes of energy carriers in terms of heat capacity per carrier C , propagation velocity v , mean free path λ , and associated relaxation time τ , where τ is equivalent to the average time between carrier collisions or scattering events. In an isotropic medium, only one of three dimensions is perpendicular to the thermal gradient; thus, only 1/3 of energy carriers are pertinent to thermal conductivity. This general expression is given in Eq. 1, which will be the basis for our MLIP-informed thermal conductivity prediction models.

$$k = \frac{1}{3}Cv\lambda = \frac{1}{3}Cv^2\tau \quad (1)$$

Building off of this framework, a versatile thermodynamic property-based kinetic theory model (KTM) was implemented by Gheribi et al. on a range of molten salt mixtures [34, 36], although the model’s lack of energy carrier property dependencies limits its theoretical interpretation. Specifically, the KTM interprets λ as the average interatomic distance obtained from the number density of the salt, but controls most of the sensitivity with composition and temperature-specific factors applied to the entire k expression rather than the individual carrier properties. This takes a functionally agnostic approach to defining energy carrier properties in favor of capturing correlations with effective thermal conductivity.

Subsequent work by Zhao et al. confined the expression of k solely to energy carrier characteristics by use of a phonon gas model [101], which considered the propagation of high-frequency vibration modes traveling at the speed of sound. Like the KTM, the phonon gas model assumed λ was equal to the average intermolecular distance within the salt. This resulted in reasonable predictions for sodium and potassium nitrate, water, and argon, while generally underpredicting halide molten salts.

Attempts to attribute the mean free path of vibrational energy carriers to the degree of short-range order in molten salts led to the recent development of a structural coherence model (SCM) of thermal conductivity [67]. The SCM resolved λ into an estimate of the structural coherence length (ℓ_{sc}) as approximated from the partial pair distribution function (pPDF), which allowed for extended mean free paths beyond the interatomic distance [67]. The SCM demonstrated increased predictability for a variety of salt types, including actinide-bearing mixtures, though it remains crucially limited by the availability of a pPDF for the particular salt composition at the relevant temperatures.

While analytical approaches leverage simplifying assumptions to provide general solutions of molten salt thermal conductivity, first-principles-informed models like the SCM are constrained by inadequately populated MD-generated pPDF data. This presents a direct parallel to the bottleneck faced by MD methods of thermal conductivity calculation, which are both time-intensive and still subject to uncertainty. To validate and implement analytical models for a lightweight parameterized formalism for predicting k , there must be a bridge between MD-generated structural information and the lower computational costs of analytical approaches.

1.3 Using MLIPS to Combine MD and SCM Approaches

The previous limiting factor for the SCM was scarcity of available pPDF data, but MLIPs create an alternative pathway for generating the required structural data. Trained on large swaths of DFT-based AIMD simulation data, MLIPs have shown promising results with near-DFT accuracy of varying elemental compositions at a fraction of the computational cost [43]. In the case of pPDF and density particularly, many MLIPs achieve as close as 0.01-0.02 Å (1-3%) for peak positions of AIMD generated distributions, and within 1-2% of AIMD predicted density [37, 75]. Because MLIPs have the potential to more rapidly generate pPDF data than with strictly AIMD, the use of the SCM model as a predictive capability to fill gaps in compositionally-varying data can be explored.

As well as producing more pPDF data, MLIPs are capable of predicting volumetric heat capacity (C_p) and adiabatic speed of sound (v_s), which opens up predictive capability for salts without available heat capacity or speed of sound data. This work presents a proof of concept for a comprehensive workflow of the SCM by simulating each property using the “SuperSalt” MLIP [75]. This provides all the parameters required to characterize energy carriers with Eq. 1. With the provision of MLIPs, this yields a self-contained thermal conductivity prediction in the energy carrier model given in Eq. 2.

$$k = \frac{1}{3}C_p v_s \ell_{sc} \quad (2)$$

The methodology of the proposed integration of the SCM and machine learning interatomic potentials is described in Section 2. Initial results over a variety of salt types and compositional studies for both monovalent and divalent salts are compared to experimental data in Section 3.

The applicability of this approach to predicting properties of salts with a large number of constituents is demonstrated for simulated high-entropy mixtures termed the “Big” 11-cation molten salt from the original SuperSalt paper [75]. The addition of an isothermal bulk modulus (K_T) prediction, and the preliminary results of applying Gheribi’s Temperature Dependence Function (GTDF) [30] to a single data point will be compared to the results of running independent MD simulations for each temperature.

Finally, the results of the integrated SCM and MLIP approach (ISM) are compared to predictions made by RNEMD performed with the same MLIP to provide a direct comparison of both accuracy and computational cost. The proposed integrated

model takes 3 hours to complete a thermal conductivity prediction, running in parallel over three H200 GPUs, while an RNEMD simulation also using the SuperSalt potential takes on average 60 hours, on a single H200 GPU (because RNEMD cannot be parallelized for a single prediction). These results support this work as a proof of concept for integrating the SCM with MLIPs to predict thermal conductivity at lower computational cost, allowing for many more predictions to be made for specific salt systems than previously possible.

2 Methodology

To leverage mechanistic analytical models with minimally time-intensive atomistic simulations, this work presents an integrated SCM—MLIP approach (ISM) for molten salt thermal conductivity characterization. This section describes the workflow and analysis process for each of the properties obtained from the implementation of MLIPs in classical MD and non-equilibrium MD simulations.

2.1 MLIP Implementation

Machine Learning Interatomic Potentials (MLIP) are large neural networks that bridge the gap between traditional AIMD (where high resolution of chemical interactions can be modeled at low-scale with extremely high computational cost) and physics-based potentials (where larger-scale systems are modeled with lower accuracy and computational cost). MLIPs can compute medium-sized systems with nearly AIMD-level accuracy, and at a fraction of the cost [43]. A previous limit to MLIPs was high training data requirements, but due to the increasing quantity of open source *ab initio* data, training MLIPs to high degrees of fidelity is becoming easier. There are a variety of MLIPs [70], with targeted-MLIPs focused on specific salt compositions, and semi-universal or universal models seeking to generalize and achieve universal applicability. A true universal model has yet to be built with high accuracy but is of high interest for future work.

The MLIP “SuperSalt” [75] is a semi-universal MLIP, with predictive capability across 11 different cation-chloride melts (Li, Na, Mg, K, Ca, Cs, Ba, Sr, Rb, Zn, Zr). SuperSalt’s architecture is a Multi-layer Atomic Cluster Expansion (MACE) neural network. Current results show that SuperSalt is capable of predicting density, bulk modulus, radial distribution functions (equivalent to pPDF), and specific heat capacity at constant pressure. Our proposed ISM model builds a workflow to predict thermal conductivity as a function of temperature by using the SuperSalt MLIP in MD simulations.

2.1.1 SuperSalt Workflow

The proposed workflow combines the SuperSalt MLIP, MD simulations, and lower-cost physics-based models to predict molten salt thermal conductivity as a function of temperature (Figure 2). Following the kinetic-theory framework of Zhao [101], the model uses MD simulations to determine the volumetric constant-pressure heat capacity (C_p), adiabatic speed of sound (v_s), and structural coherence length (ℓ_{sc}), where the

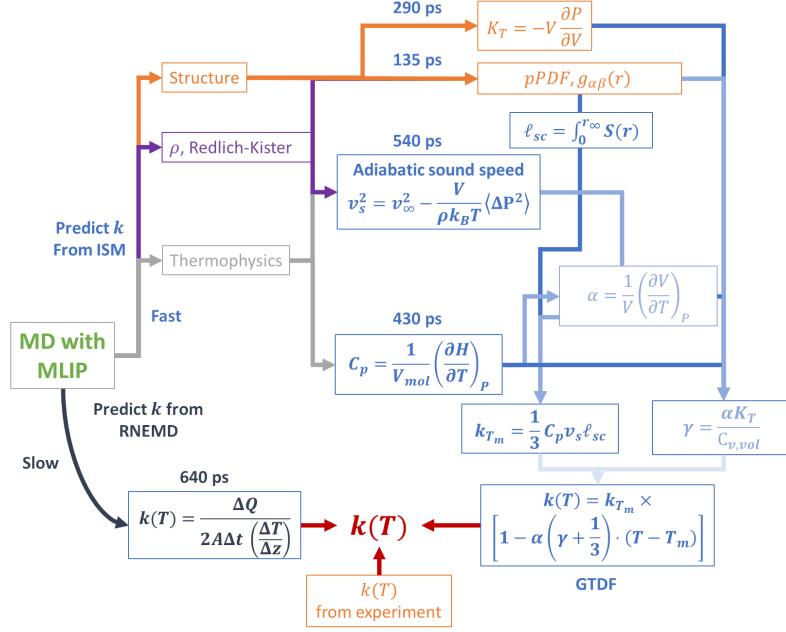


Fig. 2: Schematic of the general work flow of the integrated structural coherence and machine learning interatomic potential (ISM) model to compute different properties.

latter replaces the conventional mean free path (λ) through the structural coherence model (SCM) [67]. These properties are combined to generate a thermal conductivity prediction at the melting temperature.

To minimize computational cost, classical models are used whenever possible, including a Redlich-Kister density model [3] and Gheribi’s temperature-dependent scaling function (GTDF) [30]. The scaling function uses predicted values of the isothermal bulk modulus (K_T), together with the constant-volume heat capacity and coefficient of thermal expansion derived from the heat-capacity simulations (α), to extrapolate thermal conductivity beyond the melting temperature. This extrapolation is based on the assumption that density is the primary driver of the temperature dependence of thermal conductivity and is described in more detail in Section 2.2.7.

The first step of the workflow is to predict the density of the specific composition at its melting temperature. Accurate density values have an outsized impact on both the volumetric heat capacity at constant pressure and adiabatic speed of sound predictions [7]. Although SuperSalt has demonstrated capability in predicting density with high fidelity, an existing model that uses Redlich-Kister expansion provides an alternative, lower computational cost route, so long as sufficient experimental data exists to support the model.

Once density has been calculated, the molten salt composition (x), melting temperature (T_m), and density (ρ) are included in MD simulation scripts that generate the four main properties of interest (C_p , v_s , ℓ_{sc} , K_T) in four parallel scripts. The longest

single simulation is C_p , where enthalpy is measured at six different temperatures, and then fit with a quadratic to yield a temperature dependent value. This script allows for parallel computation of the thermal expansion coefficient, which when combined with bulk modulus and constant volume heat capacity can approximate the Gruneisen parameter which is an essential part of the GTDF [30].

The mean free path, ℓ_{sc} is calculated using the Structural Coherence Model, an algorithm that uses a pPDF to quantify the level of coherence in the molten salt [67]. Therefore, the MD simulation consists of generating an pPDF trajectory file, and analyzing the result with the SCM. Bulk modulus, K_T is calculated by measuring the volume at four different pressures and constant temperature during a separate MD simulation.

Finally, v_s is derived using Bryk’s stress fluctuation method [7], and its calculation is highly dependent on accurate density values with a propensity to statistical variance due to measurement of the stress tensor over a long time scale. Therefore, three trials of adiabatic speed of sound simulations are run, and the results are averaged.

Once C_p , v_s , ℓ_{sc} , and K_T have been predicted, an aggregate script implements Eq. 2 to predict thermal conductivity at T_m (k_{T_m}). This value is then scaled by the Gruneisen Parameter and thermal expansion coefficient to generate the GTDF ($k(T)$). This yields a thermal conductivity prediction for a 216-324 atom sized simulation space after 1105 ps of simulated time for all the constituents of k_{T_m} , followed by an additional 290 ps of simulated time to determine $k(T)$.

Because multiple independent simulations are performed, they can be run in parallel over available GPUs in a supercomputing cluster, which reduces actual computational time. The full model pipeline is designed to be run in parallel, with single user inputs of composition and temperature, and can be accessed in Section 6. Due to the small simulation size and time-scale, as well as the ability to run easily run the model in parallel, the integrated SCM-MLIP (ISM) model takes 1-2 orders of magnitude less time than a comparable RNEMD simulation (3456 atoms, 640 ps, see Section 2.3) performed with the same SuperSalt potential. In the case of 11 different compositions of the NaCl-KCl mixture, the RNEMD simulation took between 52-71 hours on a single H200 GPU, while the ISM model took between 2.5-3.5 hours on multiple H200 GPUs running in parallel, or 8.5-9.5 hours when performed on a single H200 GPU.

All MD simulations were allowed to geometrically optimize with the FIRE package after being set in a structure, using lattice substitution to reach the desired composition, and being perturbed slightly to avoid any false energy minima. The lattice substitution sets a base structure of the component with the highest oxidation number, e.g. fluorite, then replaces cations randomly in the proportion needed to reach the desired composition. Excess chlorine atoms are removed and left as vacancies in the simulation space. The code containing this method can be viewed in the GitHub repository described in Section 6.

2.2 Property Determination

This section details the process used to determine each parameter in Table 1, which presents a summary of the different parameters used in the prediction of thermal conductivity k , the dependencies and use of each parameter, and a reference to the

equation defining the parameter or model. Notational descriptions that are not obvious in Table 1 will be provided throughout Section 2.2 as the mathematics associated with property determination are described.

Table 1: Summary of model parameters, with * representing a parameter directly calculated through MD simulations.

Parameter	Symbol	Depends on	Used for	Eq.
Partial PDF*	$g_{\alpha,\beta}(r)$	V, N_α, N_β	ℓ_{sc}	(Eq. 8)
Mean free path	ℓ_{sc}	$g_{\alpha\beta}(r), S(r), \beta(r)$	k_{T_m}	(Eq. 9)
Density	ρ	Redlich-Kister	$v_s, v_\infty, V, \Delta P$	(Eq. 6)
Mol. heat capacity (const. P)*	$C_{p,mol}$	$(\frac{\partial H}{\partial T})_P$	C_p	(Eq. 3)
Vol. heat capacity (const. P)	C_p	$V_{mol}, C_{p,mol}$	k_{T_m}, α, γ	(Eq. 4)
Adiabatic sound speed*	v_s	$V, \rho, v_\infty, \Delta P$	k_{T_m}, γ	(Eq. 6)
Thermal conductivity (melt point)	k_{T_m}	C_p, v_s, ℓ_{sc}	$k(T)$	(Eq. 2)
Bulk modulus*	K_T	$V, \frac{\partial P}{\partial V}$	$\gamma, k(T)$	(Eq. 10)
Thermal expansion*	α	$V, \frac{\partial V}{\partial T}$	$k(T)$	(Eq. 5)
Gruneisen parameter	γ	α, K_T, C_p	$k(T)$	(Eq. 12)
Gheribi scaling	$k(T)$	k_{T_m}, γ, T_m	-	(Eq. 11)

2.2.1 Density

Density has a very large impact on the MLIP’s predictive performance for both constant pressure heat capacity and the adiabatic speed of sound. The effect on heat capacity is due to the conversion of molar heat capacity ($C_{v,mol}$) to volumetric heat capacity (C_p), which includes density in the denominator. The effect on adiabatic speed of sound (v_s) is due to the extremely sensitive measurements of virial stress variance, as described more fully in Section 2.2.3.

While SuperSalt is capable of predicting the density with good accuracy [75], Redlich-Kister models provide an alternative predictive route at lower computational cost, utilizing tabulated excess density terms, determined by fitting to experimental data, which account for deviations from additive molar volumes (which would otherwise predict ideal density).

Often these values are more accurate than the MD simulated values, with average error of about 2-4% [3] for most mixtures compared to 5% error by SuperSalt [75]. Because even a small error can have massive effects on both heat capacity and speed of sound, the RK model is used instead of an MD simulation for both accuracy and computational speed, whenever possible.

By starting the workflow with an RK density prediction, additional MD simulations can run in parallel without needing MD calculated density, increasing speed without sacrificing accuracy. When excess density terms are available such as those tabulated in MSD-TP [14], the Redlich-Kister model requires no GPU and adds minimal cost to the full model. When the excess terms are unavailable, calculating for density using the SuperSalt potential requires a total of 55 ps (5 ps melt, 20 ps equilibration, and 20 ps production).

2.2.2 Heat Capacity

Volumetric heat capacity at constant pressure is assumed to accurately represent an upper limit to the available transferable energy of energy carriers. In molten salts, vibrational phonons are expected to be the dominant microscopic thermal transport carrier [1, 101]. Using volumetric heat capacity as the energy of the carrier was chosen because it accounts for both optical (or transverse) and longitudinal vibrational states and thus provides an upper limit on energy [11, 15].

To calculate the molar heat capacity, $C_{p,mol}$, a 3x3x3 supercell sized simulation space is created, and once geometrically optimized, melted at 2500K for 10 ps, followed by 6 sweeps at temperatures scaled in 100K increments from the melting temperature. Each sweep consists of a 20 ps equilibration run and 50 ps production run measuring enthalpy, both in the NPT ensemble. The total computational time is 430 ps, over a simulation size between 216 and 324 atoms depending on whether the mixture is predominantly comprised of monovalent or divalent cations. Taking the derivative of the change in enthalpy with temperature yields molar heat capacity at constant pressure:

$$C_{p,mol} = \left(\frac{\partial H}{\partial T} \right)_P \quad (3)$$

Volumetric heat capacity C_p , is calculated by dividing the molar heat capacity $C_{p,mol}$ by the volume per mole of the simulation space V_{mol} :

$$C_p = \frac{C_{p,mol}}{V_{mol}} \quad (4)$$

The coefficient of thermal expansion, α , is found by scaling the change in volume with temperature by volume, in this form:

$$\alpha = \frac{1}{V} \left(\frac{\partial V}{\partial T} \right)_P \quad (5)$$

2.2.3 Adiabatic Speed of Sound

Adiabatic speed of sound is assumed in previous kinetic theory models to accurately represent the average group velocity of energy carriers [67], although there is still some uncertainty surrounding observed “fast sound” in measured dispersion relations [16–18, 21, 38, 54]. For the scope of this model, it is assumed that the group velocities of the dominant energy carriers are equivalent to the speed of sound v_s .

Measuring adiabatic speed of sound in molten salts is incredibly difficult, due to temperature limits of conventional transducers and the complexity associated with using a buffer rod between the transducers and fluid being tested [23, 41, 56]. As a result, many speed of sound predictions are done by computational rather than experimental means. The two previous computational methods are the Dynamic Structure Factor and Thermodynamic derivation. While both the Dynamic Structure Factor and Thermodynamic methods are used widely in literature, and sometimes in tandem to

verify results [52], they both have characteristics that make them incompatible with the ISM model's desired low computational cost.

In order to circumvent the difficulties of calculating adiabatic speed of sound by simulating spatial wave propagation over a large size, or thermodynamic properties over a long timescale, Bryk et al. proposes a model driven by stress fluctuation [7]. The overall derivation of his prediction is as follows:

$$v_s^2 = v_\infty^2 - \frac{V}{\rho k_B T} \langle \Delta P^2 \rangle \quad (6)$$

where v_s is the adiabatic speed of sound, v_∞ is the high-frequency speed of sound, and $\langle \Delta P \rangle$ is the variance of virial stresses. Essentially, Bryk models the adiabatic speed of sound as a balance between a hypothetical high-frequency limit, v_∞ , and the degree of structural relaxation in the liquid. The effect of relaxation is represented by the variance of virial stress scaled by volume, density, Boltzmann constant, and temperature.

The term v_∞ , or high-frequency speed of sound, is calculated by measuring the longitudinal mass current $J(q, t)$, and solving for the long wavelength limit of the ratio between the variance of mass current's time derivative $\dot{J}(q, t)$ to the variance of $J(q, t)$.

The fluctuation term $\frac{V}{\rho k_B T} \langle \Delta P^2 \rangle$ is calculated by measuring the longitudinal stress vector tensors σ_{xx} , σ_{yy} , and σ_{zz} , and scaling the square of the variance of those tensors $\langle \Delta P^2 \rangle$ over the simulation time by V , ρ , k_B , and T . $\langle \Delta P^2 \rangle$ is derived in the following form:

$$\langle \Delta P^2 \rangle = \frac{\langle \Delta \sigma_{xx}^2 \rangle + \langle \Delta \sigma_{yy}^2 \rangle + \langle \Delta \sigma_{zz}^2 \rangle}{3} \quad (7)$$

The simulation is run in a 3x3x3 supercell (216-324 atoms) where after a 10 ps melt with a Langevin thermostat in NVT ensemble, a 20 ps equilibration phase is run with a Bussi CSVR thermostat. The Bussi CSVR thermostat was selected in order to prevent any flying ice cube effects [4]. After the CSVR equilibration, a NVE equilibration runs for 80 ps followed by a 70 ps production run where the stress fluctuation is recorded. The mass current, time derivative of the mass current, and the fluctuation term are recorded once every 10 time steps of the production run, resulting in 7000 total data points.

Bryk's method elegantly avoids the difficulties of spatial and temporal scale associated with simulating wave propagation or thermodynamic properties. This enables the ISM model to generate speed of sound approximations comparable to literature values, over an extremely small time scale (180 ps) and simulation size (216-324 atoms). Due to statistical variance across different trajectory seeds, the simulation is repeated with identical physical lattice seeds but randomized trajectory seeds, and then averaged to yield v_s in the kinetic theory model (Eq. 2). After extensive trial and error, the optimal number of trials was set at three to preserve computational speed and maximize accuracy (There are cases where three does not appear to be sufficient, as discussed in Section 4.1). As a result, the total computational cost of approximating the adiabatic speed of sound is 540 ps.

2.2.4 Partial Pair Distribution Function

The pair distribution function (PDF), denoted by $g(r)$, is a form of pair correlation function corresponding to the likelihood of any two atoms being separated by a distance r , relative to the probability of a completely random distribution (as in an ideal gas) [84]. The partial pair distribution function (pPDF) represents the probability of finding specific ions α and β separated by distance r , designated by $g_{\alpha\beta}(r)$. In the context of MD simulations, the pPDF is calculated as a normalized histogram of pair separation distances between ions α and β , with a delta function wherever $r = r_{ij}$:

$$g_{\alpha\beta}(r) = \frac{V}{N_{\alpha}N_{\beta}} \left\langle \sum_{i=1}^{N_{\alpha}} \sum_{j=1}^{N_{\beta}} \delta(r - r_{ij}) \right\rangle \quad (8)$$

where V is the volume of the system, N_{α} and N_{β} are the respective numbers of ions α and β , and the bracket notation indicates the ensemble average taken over multiple simulation frames.

The pair distribution function provides a time-averaged snapshot of the average short-range ordered structure as it is dictated by the intermolecular potentials of the component salt ions. This allows the $g_{\alpha\beta}(r)$ to encode the effect of the ionic environment on the liquid structure. Ions oscillate about semi-equilibrium positions above the Frenkel frequency, which enables the approximation of a static structure for vibrational energy carriers if the pertinent time domain is well below the relaxation time of the liquid configuration. Because vibrational energy transfer is expected to form the dominant mode of thermal conduction [12, 87], the pPDF has been used to predict the mean free path of vibrational energy carriers in terms of the decay of coherent structure to the degree of its deviation from an ideal crystal [67]. This method is discussed in Section 2.2.5.

The pPDF is obtained by setting a 3x3x3 supercell and running a 10 ps Langevin melt, 25 ps equilibration under Nosé-Hoover thermostat and NPT ensemble, and a 100 ps production run with Langevin thermostat and NVT ensemble. This matches the SuperSalt simulation which allowed validation between the ISM model results and published results of the same potential. The total computational cost is 135 ps for a 216-324 atom-sized space.

2.2.5 Structural Coherence Length

In previous models that utilize the general kinetic theory representation of thermal conductivity, both Zhao [101] and Gheribi [36] assume the mean free path to be the average interatomic distance between atoms as determined by the number density at melting temperature T_m . Assuming that the heat capacity and speed of sound accurately capture energy carrier available energy and average velocity, it is this assumption that leads to consistent underprediction in the case of Zhao’s phonon gas model, with Gheribi’s kinetic theory relying heavily on compensating prefactors to limit the underprediction. Because volumetric heat capacity can be predicted for most salts with high accuracy and acts as an upper limit for the transferable energy available from phonons [15], it can be inferred that adiabatic speed of sound and

assumed mean free path are usually the cause for underpredictions. For salts with reliable experimentally derived values of adiabatic speed of sound, underprediction by a predictive model indicates that the source of deviation is the mean free path. This hypothesis is explored more fully in Section 3.

The structural coherence length, ℓ_{sc} , represents the degree to which coherence via vibrational transfer is maintained in the medium, compared to an ideal coherence of alternating cation-anion chains that have a high bond strength and no phonon scattering. Contributions to decoherence are interpreted to result principally from three disruption factors (b_i)—the probability of non-ideal recipients in a cation-anion chain (b_{NI}), weak bond strength (b_{KF}), and incoherent phonon transfer (b_{PH})—approximated on a scale from 0–1 using key features of the pPDF [67]. These are combined into a decoherence function $\beta(r)$, calculated by $\beta(r) = \sum_i b_i(r) / [1 - b_i(r)]$ over the range of the pPDF. This characterizes an energy carrier survival function $S(r)$, from which the structural coherence length at T_m can be determined as given in Eq. (9).

$$\ell_{\text{sc}} = \int_0^{r_\infty} S(r) dr = \int_0^{r_\infty} \exp\left(-\int_0^r \beta(r') dr'\right) dr \quad (9)$$

Because the SCM is a simple numerical algorithm as opposed to a particle-tracking MD simulation, it adds negligible computational cost to the ISM model. The pPDF and ℓ_{sc} using this approach are given for the cation-anion pairs in KCl in Figure 3a and for NaCl–KCl (50-50 mol%) in Figure 3b. These are compared to pPDF data generated by a polarizable-ion model for KCl [95] and a rigid-ion model for NaCl–KCl. Though peak height differs somewhat between the models in both cases, the peak radial position differs by less than 5%, with less than 2.4% deviation in the calculated ℓ_{sc} . The significant drop in $S(r)$ at the first peak position in the NaCl–KCl mixture as compared to the pseudo-unary KCl stems from the phonon transfer factor b_{PH} , which predicts a total disruption of coherent vibrational energy transfer proportional to the composition of dissimilar ions. This is derived from the simplified assumption that distinct resonant frequencies formed by differing ion masses and bond strengths cannot couple [67]. Insights and limitations involved in this assumption are discussed in Section 3.

2.2.6 Isothermal Bulk Modulus

The isothermal bulk modulus of a material represents the compressibility at a certain temperature and is calculated by measuring the change in volume across changing pressure.

$$K_T = -V \frac{\partial P}{\partial V} \quad (10)$$

The ISM model predicts K_T by simulating a 3x3x3 supercell, melting it with a Langevin thermostat in NVT ensemble for 10 ps, then cooling to the target temperature and equilibrating under the NPT ensemble for 20 ps. Once equilibrated at the temperature, the melt is equilibrated at the first pressure for 20 ps, and data is collected for 50 ps in the production run. This is repeated for a total of 4 pressures. The total computational cost of this property calculation is 290 ps of a 3x3x3 simulation space (216-324 atoms).

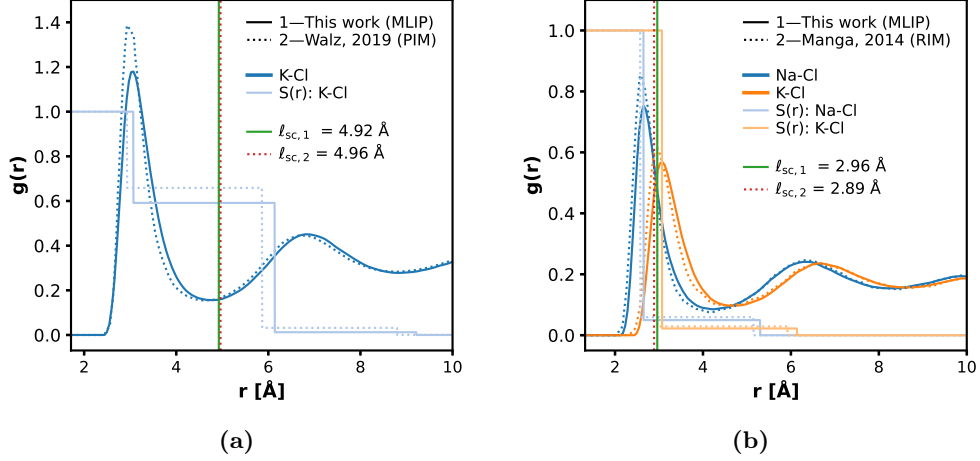


Fig. 3: Comparisons of the partial pair distribution function (pPDF) for cation-anion pairs in (a) KCl and (b) NaCl–KCl (50-50 mol%) from SuperSalt MLIP as represented by solid lines. Dotted lines represent pPDFs from (a) the polarizable-ion model (PIM) used by Walz [95] and (b) the rigid-ion model (RIM) by Manga [53]. The survival function $S(r)$ is also depicted, with vertical lines representing the structural coherence length ℓ_{sc} as calculated from this work ($\ell_{sc,1}$) and from pPDFs in literature ($\ell_{sc,2}$).

Simulating the isothermal bulk modulus, K_T , allows for the Gruneisen parameter γ to be calculated using the C_p and α values, allowing us to apply Eq. (11) (to be described in the next section) to a single thermal conductivity prediction at T_m and extrapolate for higher temperatures.

2.2.7 Gheribi’s Temperature Dependence Function (GTDF)

It is proposed by Gheribi et al. that thermal conductivity decreases with increasing temperature in a linear fashion and that the dominant contributor to this negative correlation is temperature-dependent density. Although the available energy and speed of atomic motion increase with temperature, the average interatomic spacing increases as density decreases. This reduces the volumetric specific heat and the velocity of energy carriers, resulting in an overall decrease in thermal conductivity that is well corroborated across most computational predictive methods [15]. This also matches existing experimental data collected by established methods [63, 80, 85].

Gheribi’s derivation of this relationship uses the thermal expansion coefficient α as well as the Gruneisen parameter γ in the following form:

$$k(T) = k_{T_m} \left(1 - \alpha \left[\gamma + \frac{1}{3} \right] \cdot (T - T_m) \right) \quad (11)$$

where γ can be derived using the constant-volume heat capacity C_v , bulk modulus K_T , and α in the following form:

$$\gamma = \frac{\alpha K_T}{C_v} \quad (12)$$

which assumes that constant-volume heat capacity can be approximated as:

$$C_v = C_p - T_m \alpha^2 K_T \quad (13)$$

By predicting the bulk modulus as well as the three properties required for the SCM, a single data point for thermal conductivity can be extrapolated as a function of temperature. However, due to the many substitutions used to calculate γ , this extrapolation is highly dependent on the accuracy of the K_T and α approximations. K_T is used twice and α three times, meaning even slight errors in their predictions are heavily amplified.

Addition of this function to the ISM model requires simulation for K_T and therefore adds 290 ps to the computational cost. However if the theorized temperature dependence is validated, it will result in a net reduction to computational cost. The preliminary results of the function compared to running the simulation at multiple temperatures are discussed in Section 3.5.

2.3 Thermal Conductivity using RNEMD with SuperSalt Potential

A commonly used type of the non-equilibrium methods, reverse non-equilibrium molecular dynamics (RNEMD), operates by simulating heat flux and recording changes in temperature [61]. This is achieved by dividing the simulation space into small slices, or “bins,” then creating a temperature gradient by setting cold and hot zones. A bin at one end is set as cold, the bin in the middle is set as hot, and periodic boundary conditions are applied. To simulate the temperature gradient, the cold and hot zones are periodically scanned at a set time interval. The atom with the highest velocity in the cold bin and the atom with the lowest velocity in the hot bin are identified, then switched instantaneously by replacing their velocity vectors with each other. This allows for momentum and kinetic energy to be conserved, which pairs cleanly with the NVE, microcanonical ensemble [44] where particles (N), the system’s volume (V), and the total energy (E) are held constant.

Once the temperature gradient is established, the simulated heat flux works against the fluid’s intrinsic conductivity, and thermal conductivity can be calculated by integration of Fourier’s law. Total thermal conductivity, k , is set equal to total kinetic energy moved by the RNEMD velocity swaps, ΔQ , divided by twice the cross sectional area, A because of the bi-directional flow of heat with the hot zone placed between cold zones, the total time interval Δt , and the spatial temperature gradient $\Delta T/\Delta z$:

$$k = \frac{\Delta Q}{2A\Delta t(\frac{\Delta T}{\Delta z})} \quad (14)$$

The RNEMD method has a significant advantage over both Green-Kubo and NEMD methods because the simulated heat flux is explicitly known. This eliminates the complexity associated with partial enthalpies calculations and the need to derive a field that simulates a temperature gradient. By calculating thermal conductivity using only kinetic energy and local temperature, RNEMD positions itself as a uniquely compatible candidate for integration with MLIPs. This is because current MLIPs struggle with to accurately capture stress tensors but easily and accurately measure kinetic energy and temperature. This simplicity means that it also converges much more quickly than the Green-Kubo method, with similar accuracy [15].

The largest weakness of the RNEMD method is the impact of finite-size effects. The dominant form of heat transfer in molten salts is assumed to be from vibrational phonons, based on the phonon theory of liquid thermodynamics where the phonons are eigenstates of topologically disordered liquids instead of harmonic vibrations in a structured crystal lattice [1]. If the distance between the hot middle slabs and cold end slabs is much larger than the mean free path of the phonon, boundary slabs truncate the vibrational modes and artificially lower the simulation’s thermal conductivity prediction ([10, 78, 82]).

For this study, the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) was used to apply RNEMD. LAMMPS was compiled for CUDA gpu support through the Kokkos package. The SuperSalt potential was used through the MACE pair style with no domain decomposition.

A 6x6x12 supercell was created with periodic boundaries. It was structured as a NaCl crystal and the sodium atoms were replaced with potassium according to the desired concentration for the simulation. In the NPT ensemble, the cell was overheated to twice the target temperature for 20 ps to achieve melting, cooled to the target temperature for 20 ps, and allowed to equilibrate at the target temperature for 200 ps. RNEMD was then applied for 400 ps. The LAMMPS thermal conductivity fix divided the supercell into 20 bins, with bin 1 assigned to be the cold bin and bin 11 assigned to be the hot bin. Then, in the NVE ensemble, the highest velocity atom in the cold bin and the lowest velocity atom in the hot bin had their velocity vectors switched every 20 timesteps while the total kinetic energy transferred is tracked. Thermal conductivity of the salt was then calculated according to Eq. (14).

3 Results

This section will present the results for pseudo-binary (NaCl-KCl and MgCl₂-KCl) salts at varying molar fractions of each unary salt, as well as the results for the eutectic compositions of the pseudo-binary MgCl₂-KCl and pseudo-ternary NaCl-KCl-MgCl₂ salts. The impact of supercell size will be investigated using a high entropy, 11 cation salt, with additional analysis for high entropy salts provided in the Supplementary Materials. Finally, we will investigate the validity of the Gheribi temperature dependent function (GTDF) by comparing it to results of directly applying the ISM model at multiple temperatures and experimental data.

3.1 ISM vs Previous SCM Predictions

Table 2 presents the predicted values of C_p , v_s , ℓ_{sc} , and k based on the ISM method where all data was based on MD simulations. These predictions are compared to the original predictions for thermal conductivity by the SCM approach [67], where predictions of k could only be made where there was published data. In addition to the pPDFs obtained from literature, the first implementation of the SCM used experimental values of heat capacity and sound velocity. Thermodynamic mixing rules, also used with the KTM [29], were applied when experimental values for the particular salt mixture did not exist. This is in contrast to the ISM model, which predicts all these properties (C_p , v_s , and ℓ_{sc}) directly using the SuperSalt MLIP as the basis for MD simulations. The differences between the C_p and ℓ_{sc} values for the ISM and SCM approaches are relatively low compared to the differences observed in v_s , with Mg-bearing mixtures exhibiting the largest differences. The potential reasons for this distinct underestimation will be discussed in Section 4.1.

Table 2: Thermal conductivity k , volumetric heat capacity C_p , adiabatic speed of sound v_s , and mean free path ℓ_{sc} predictions of several salts at their melting temperature (T_m) from the proposed ISM method compared to the literature-obtained values described in the original SCM paper [67]. Input parameters for unary endmembers and mixtures were obtained from [5, 8, 19, 36, 39, 42, 45, 57, 71, 74, 91]. Compositions are in molar percentages, and **bolded** salts represent those with detailed results presented in this work.

Salt Composition (mol %)	T_m [K]	C_p [MJ m ⁻³ K ⁻¹]			v_s [m s ⁻¹]		
		ISM	SCM	%Δ	ISM	SCM	%Δ
Unary							
LiCl	883	2.36	2.30	-2.6	2079.5	1855.8	10.8
NaCl	1074	1.79	1.85	-3.2	1803.1	1773.6	1.7
KCl	1043	1.52	1.51	0.5	1863.3	1632.7	14.1
MgCl ₂	987	1.69	1.63	3.7	1003.0	2101.0	-52.3
CaCl ₂	1045	1.86	1.93	-3.6	1999.3	2065.0	-3.2
SrCl ₂	1146	1.78	1.80	-1.1	1959.3	1873.1	4.6
Binary Mixtures							
LiCl-KCl (50-50)	738	2.05	1.99	3.0	1906.5	2004.7*	-4.9
LiCl-KCl (63.7-36.3)	665	2.19	2.20	-0.4	1831.2	2109.6*	-13.2
NaCl-KCl (50-50)	930	1.67	1.73	-3.5	1959.3	1799.6*	3.0
LiCl-CaCl ₂ (70-30)	771	2.24	2.23 *	0.5	1598.5	2101.3*	-23.9
NaCl-CaCl ₂ (49.03-50.97)	779	2.16	2.23	-3.3	2001.0	2040.0*	-1.9
KCl-CaCl ₂ (71.8-28.2)	921	1.80	1.76	2.3	2020.4	1788.9*	12.9
Ternary Mixtures							
NaCl-KCl-MgCl ₂ (33-22-45)	658	2.03	1.92	5.2	1280.2	2082.8*	-38.5
NaCl-KCl-MgCl₂ (21-41-38)	660	1.89	1.93	-1.9	1571.7	2052.6*	-23.4
NaCl-KCl-CaCl ₂ (41.7-5.8-52.5)	769	2.03	2.02 *	0.5	1822.1	2029.7*	-10.2
NaCl-MgCl ₂ -CaCl ₂ (53.5-31.5-15)	829	2.08	1.95	6.5	1446.4	2039.0*	29.1
Salt Composition (mol %)	ℓ_{sc} [Å]			k [W m ⁻¹ K ⁻¹]			
	ISM	SCM	%Δ	ISM	SCM	Exp	%Δ SCM/Exp
Unary							
LiCl	3.95	4.26	-7.5	0.576	0.681	0.626	-8.0
NaCl	4.45	4.54	-2.1	0.512	0.498	0.519	-1.3
KCl	4.92	5.10	-3.5	0.427	0.389	0.419	9.8
MgCl ₂	4.54	4.54	0.0	0.330	0.518	0.553	-40.3
CaCl ₂	4.69	4.76	-1.3	0.582	0.631	0.853	-31.7
SrCl ₂	4.90	5.06	-3.1	0.570	0.569	0.720	-20.9
Binary Mixtures							
LiCl-KCl (50-50)	2.82	2.89	-2.3	0.367	0.359	—	—
LiCl-KCl (63.7-36.3)	2.99	2.81	6.6	0.400	0.434	0.626	-36.1 [†]
NaCl-KCl (50-50)	2.89	2.87	2.9	0.324	0.299	0.448	-27.7
LiCl-CaCl ₂ (70-30)	2.85	2.77	2.9	0.340	0.432	—	—
NaCl-CaCl ₂ (49.03-50.97)	2.86	2.82	1.5	0.411	0.434	0.583	-29.4
KCl-CaCl ₂ (71.8-28.2)	3.38	4.14	-18.4	0.420	0.420	—	—
Ternary Mixtures							
NaCl-KCl-MgCl ₂ (33-22-45)	2.74	2.65	3.4	0.237	—	—	—
NaCl-KCl-MgCl₂ (21-41-38)	2.48	2.50	2.8	0.248	0.367	—	—
NaCl-KCl-CaCl ₂ (41.7-5.8-52.5)	2.86	2.80	2.3	0.353	0.382	—	—
NaCl-MgCl ₂ -CaCl ₂ (53.5-31.5-15)	2.40	2.51	-4.3	0.277	0.333	0.459	-39.7

* Estimated using thermodynamic mixing rules from endmember properties.

[†] Experimental composition differed from PDF model used by more than 15% for at least one endmember.

3.2 Compositional Variation Studies

3.2.1 Monovalent (NaCl-KCl)

NaCl-KCl was selected as the subject of a compositional variation application of the model due to its widely documented properties as a heat transfer salt for concentrated solar power [28] and because it was included in a recent round robin study [55]. Because the monovalent ions are less polarizable than ions like Be^{2+} or Mg^{2+} [79, 90], the classical potential nonpolarized Born–Mayer–Huggins–Fumi–Tosi (BMHFT) [27, 86] may be used, which runs significantly faster compared to AIMD-based potential calculations. As a result, there is a high quantity of available predicted data for varying compositions of NaCl-KCl. In Figure 4a, ISM model predictions are compared to RNEMD predictions at 1100K, based on literature values using the BMHFT potentials [20, 50] and new calculations from the current work that used the SuperSalt MLIP (ISM). They are also compared to experimental measurements [64, 72].

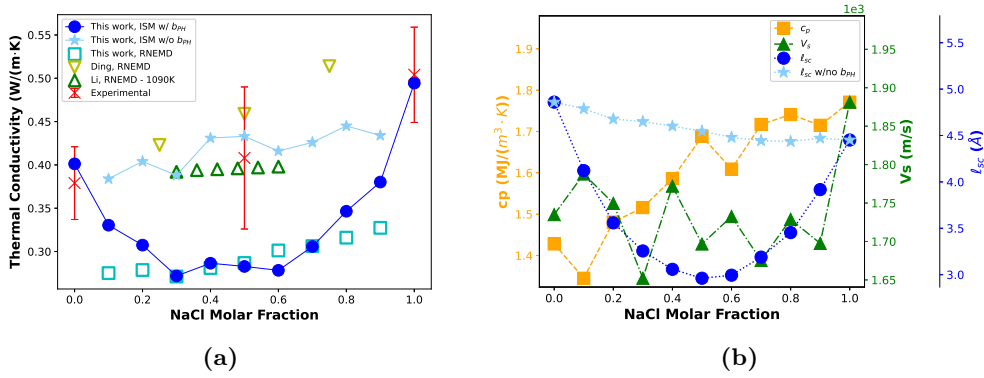


Fig. 4: (a) The full compositional plot of NaCl-KCl, including ISM results with and without the phonon transfer factor b_{PH} , RNEMD predictions with the SuperSalt potential, RNEMD predictions with the BMHFT potential [20, 50], and experimental values. Endmember (NaCl and KCl) experimental measurements are from forced Rayleigh scattering [64], and the 0.5NaCl-0.5KCl measurement is from a modified transient hot-wire needle probe [72]. (b) The three parameters used to calculate k : C_p , v_s , and ℓ_{sc} , plotted with changing composition.

The results are noticeably different at mixed salt compositions, where the ISM begins to predict significantly lower k compared to both experimental measurements and RNEMD performed with BMHFT potentials. When the individual parameters are isolated (Figure 4b), it is evident that the main cause is in the ℓ_{sc} values, in which the phonon transfer factor b_{PH} is the most influential factor in mixtures. To illustrate this, the ISM prediction is also shown where b_{PH} is not included (ISM w/o b_{PH} in Figure 4b). While this term improves predictions for pseudo-unary salts and most fluorides and actinide-bearing mixtures, in the case of NaCl-KCl, it appears to underestimate the degree of phonon transfer. This is explored more deeply in Section 4.2.

The compositional variation in ℓ_{sc} as calculated using pPDF data shows a low-noise trend, indicating the stability of pPDF predictions. The variation in C_p and v_s exhibits more scatter, observed in the overall predictions of k . The noise suggests that the observed minimum at 0.3NaCl-0.7KCl is likely due to a low v_s prediction rather than representing a true minimum in k .

Figure 5 presents the results of the ISM model (using the b_{PH} factor) for varying compositions of NaCl-KCl as functions of temperature. The GDTF (Eq. 11) model was used to extrapolate $k(T)$ from the thermal conductivity predicted at the melting temperature (k_{T_m}) and showed that both k_{T_m} and the slope of the GDTF are composition dependent.

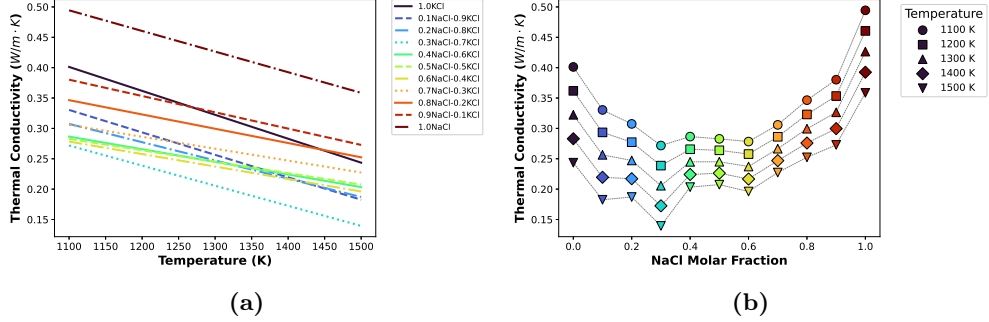


Fig. 5: (a) Thermal conductivity as a function of temperature for NaCl-KCl, from 0 to 100% molar fractions, in 10% intervals. Each composition is extrapolated out from 1100K. (b) NaCl-KCl thermal conductivity predictions at 1100K, from 0-100% molar composition, scaled up using the GTDF in 100K intervals. It is unclear whether the dip is reflective of the salt's actual properties or an error by the model. The colors for each line in (a) match markers of the same composition in (b).

Of particular note in this study was the difference in computational cost between the integrated SCM-MLIP (ISM) model and the traditional RNEMD method when both were run using the SuperSalt potential. While the RNEMD approach was only 640 ps long compared to the total 1105 ps of the ISM model, its simulation size was 3456 atoms compared to 216 atoms. This resulted in 52-71 hours (60.5 hours average) of actual runtime compared to an average of 3 hours of actual runtime by the ISM model. This demonstrates the degree to which the integrated SCM-MLIP model reduces the computational cost of thermal conductivity predictions.

3.2.2 Divalent ($MgCl_2$ -KCl)

The second compositional variation study examined $MgCl_2$ -KCl. Molten salt mixtures containing Mg^{2+} are known to exhibit networks or cation chains that form due to their high polarizability [24], which makes them incompatible with BHMFT potentials [96]. As a result, Mg^{2+} containing mixtures require full AIMD simulation, introducing

additional difficulties for predicting certain properties due to larger time and simulation size demands. These mixtures thus exhibit a unique need for MLIP development, where AIMD accuracy can be maintained as simulation times and the number of atoms are increased. Recent developments include specific potentials developed for the $\text{MgCl}_2\text{-KCl}$ eutectic [49] and for $\text{SrCl}_2\text{-KCl-MgCl}_2$ [100].

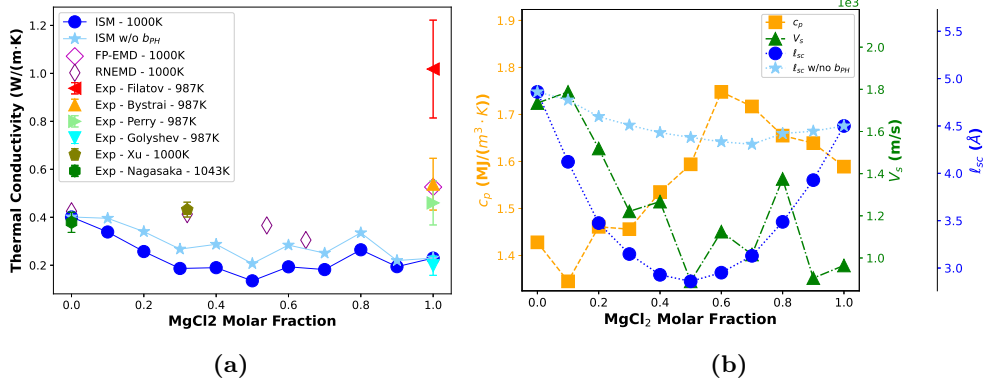


Fig. 6: (a) $\text{MgCl}_2\text{-KCl}$ thermal conductivity ISM, EMD [33], and RNEMD [51] predictions at 1000K, from 0-100% molar composition, compared to experimental data at 987 K from Filatov [26], Bystrai [2], Perry [35], and Golyshev [31], and 1000K from Xu [98], with 1043K from Nagasaka [64]. Cited errors are: $\pm 7\%$ for Xu and $\pm 11\%$ for Nagasaka. All other experimental data is estimated to be $\pm 20\%$. (b) The three properties used to calculate k : C_p , v_s , and ℓ_{sc} , plotted with changing composition.

Using the SuperSalt MLIP, Figure 6 presents the results of the compositional study (ISM, with and without the b_{PH} factor), with experimental data points plotted simultaneously for reference ([2, 26, 31, 35, 64, 98]). In the case of 0.8 MgCl_2 -0.2 KCl , the observed local maximum is likely due to a high v_s prediction rather than a true maximum. While the value of ℓ_{sc} as calculated using pPDF data and without the b_{PH} factor shows a clean trend, C_p and v_s show more scatter than observed with the NaCl-KCl mixture, which affects the overall predictions of k . However, a downward trend in v_s can be seen with $\text{MgCl}_2\text{-KCl}$ but is absent in the NaCl-KCl mixture. Combined with the upward trend in C_p as a function of increasing MgCl_2 (Figure 7b), these two parameters seem to interact with ℓ_{sc} in such a way as to have less deviation in the predicted k compared to NaCl-KCl , either with or without the inclusion of the b_{PH} factor. This demonstrates the relevance of energy carrier models, in which contributions to k can begin to be distinguishable.

Figure 7 shows the impact of the GTDF on the salt, where small molar fractions of MgCl_2 are predicted to have a stronger temperature dependence on $k(T)$ than MgCl_2 rich salts. Based on Eq. 11, this implies that there is less variability in K_T and α due to the MgCl_2 . To determine if this weak temperature dependence predicted

by the GTDF is valid, we will next investigate the predicted $k(T)$ values directly by implementing the ISM model in the next section for eutectic compositions.

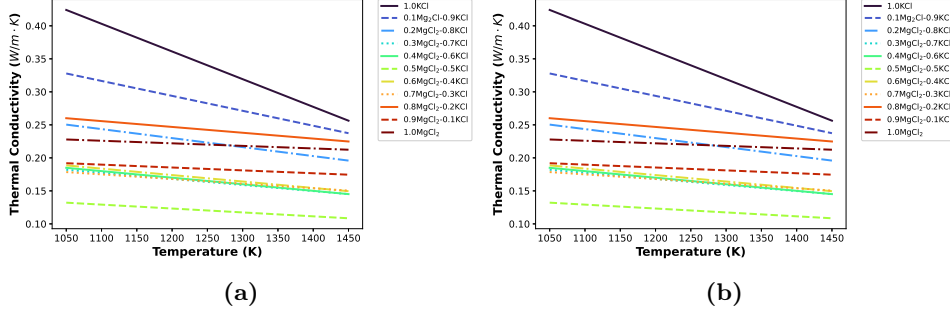


Fig. 7: (a) Thermal Conductivity as a function of temperature for $MgCl_2$ -KCl, from 0—100% molar fraction, in 10% intervals. Each composition is extrapolated out from 1050K, because KCl melts at 1044K. (b) $MgCl_2$ -KCl thermal conductivity predictions at 1050K, from 0-100% molar composition, scaled up using the GTDF in 100K intervals. As the percentage of Mg increases, the temperature dependence decreases significantly, with predictions clustering more closely together.

3.3 $MgCl_2$ -KCl and NaCl-KCl- $MgCl_2$ Eutectic Prediction Comparisons to Experimental Values

To explore the local minima observed with increasingly complex compositions, the integrated model predictions were compared to experimental measurements of the eutectic mixtures of $MgCl_2$ -KCl (0.32-0.68 mol%) and NaCl-KCl- $MgCl_2$ (0.2046-0.4131-0.3823 mol%).

Figures 8a and 8b presents the predicted thermal conductivity of the eutectic salts as functions of temperature ($k(T)$) by direct implementation of the ISM method at each temperature and by the extrapolation of thermal conductivity at the melting temperature (k_{T_m} using the GTDF. Each plot shows data with and without the phonon transfer factor b_{PH} in the calculation of ℓ_{sc} to investigate the potential impact of the decreased structural coherence length on model accuracy. Removal of b_{PH} results in closer agreement between the ISM and Xu’s experimental data in Figure 8a [97]. Xu’s measurement was performed using the laser flash analysis (LFA), which can overpredict thermal conductivity if all heat transfer modes aren’t fully accounted for [58] but has proven to provide data with a negative temperature dependence for chloride salts [9].

Figure 8c contains the pPDF of KCl- $MgCl_2$ (0.68–0.32 mol%), with the survival function $S(r)$ and ℓ_{sc} calculated with and without the phonon transfer factor b_{PH} , the exclusion of which results in a 51% increase in ℓ_{sc} . Increases in ℓ_{sc} were also observed with the pseudo-ternary mixture when the b_{PH} factor is not considered (Figure 8d). The validity of this term is proposed to be dependent on the degree to which disparate

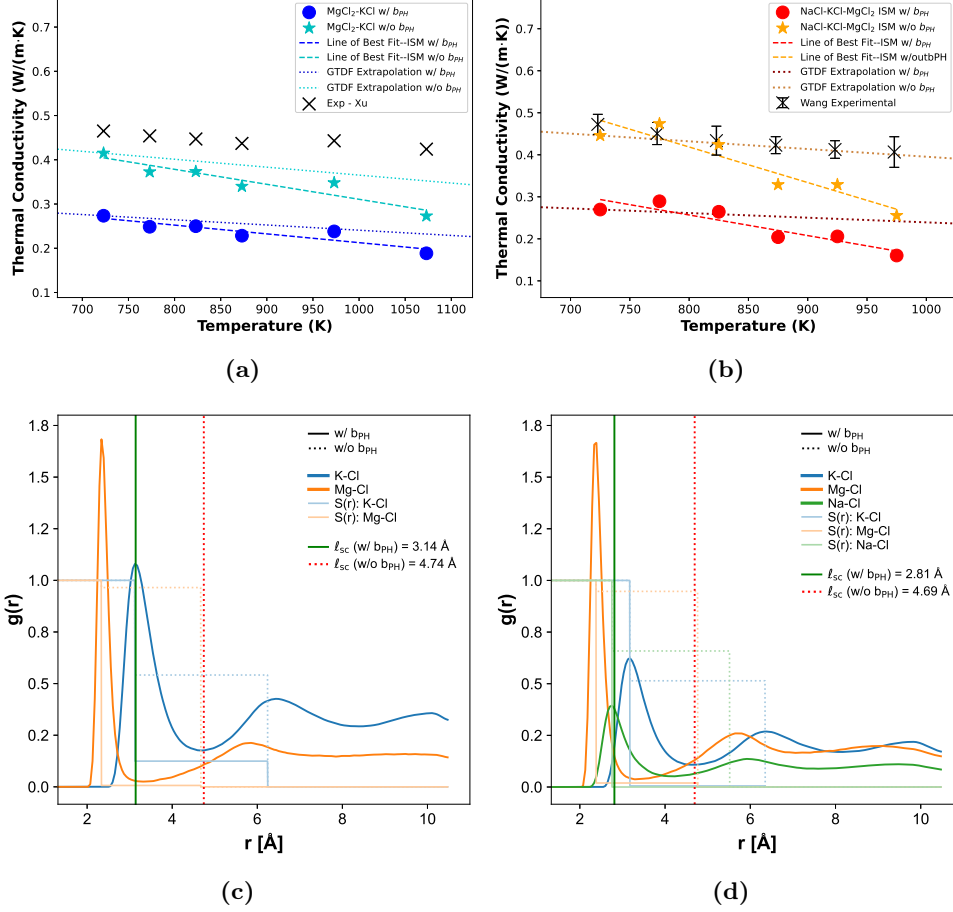


Fig. 8: (a) The ISM predictions of MgCl₂-KCl eutectic over 6 temperatures, compared to the experimental values by Xu [97], and the predictions of the ISM with the removal of the b_{PH} Factor. (b) Model predictions for the NaCl-KCl-MgCl₂ eutectic at 6 discrete temperatures, compared to experimental measurements by Wang [94] with LFA. ISM predictions with the removal of the b_{PH} term are also plotted, and show closer agreement to experimental data than observed in the MgCl₂-KCl eutectic. Plot of pPDF of (c) KCl-MgCl₂ (0.68–0.32 mol%) and (d) NaCl-KCl-MgCl₂ (0.205–0.413–0.382 mol%) and calculated ℓ_{sc} with and without inclusion of the b_{PH} factor.

cation species enable coherent vibrational energy transfer. If correct, an underestimation of b_{PH} may indicate that the salt exhibits a greater capability for vibrational energy propagation than the current discrete values of the b_{PH} currently allows. This is partially supported by the higher predicted value of the ternary than binary salt and the greater agreement of the ternary prediction with measured properties.

3.4 11-Cation SuperSalt Melt

The ability to predict thermophysical properties for salts with many components is of particular interest for the design of liquid-fueled MSRs, because fission products change the fluid composition over time. In the original SuperSalt paper, predictions for heat capacity and density were given for a 11-cation component salt termed “Big” salt [75]. The 11-cation molten salt in SuperSalt, $\text{Li}_2\text{Na}_4\text{Mg}_3\text{K}_5\text{Ca}_9\text{ZnRb}_3\text{Sr}_2\text{Zr}_6\text{Cs}_2\text{Ba}_2\text{Cl}_{74}$, shows the ability of the machine learning interatomic potential (MLIP) to simulate highly disordered mixtures at AIMD level accuracy.

This hypothetical salt was simulated using the ISM, with an adjustment to the initial simulation size in order to more accurately capture contributions of trace cations. Instead of a $3\times 3\times 3$ supercell of 313 atoms, a $4\times 4\times 4$ supercell of 741 atoms was used for the MD simulations which were conducted with the same automated scripts as all other molten salts in this study. Despite the complexity of this mixture, ISM predictions did not take more time compared to the unary or eutectic systems when performed with a $3\times 3\times 3$ sized simulation, indicating that computational cost of the ISM is independent of salt complexity. Both density and specific heat values predicted by the ISM model agree with the published values in the SuperSalt paper [75] within 1%, which strengthens confidence in the reliability of MD simulation performed using the automated pipeline of the ISM.

The predicted thermal conductivity also appears plausible, although the spread is large and due to high variance of v_s (Figure 9). This indicates that for highly complexed salts, more than three simulations under the same initial conditions used to predict v_s may be needed. The values themselves are also likely underestimates due to the effect of MgCl_2 on v_s , which is discussed in Section 4.1.

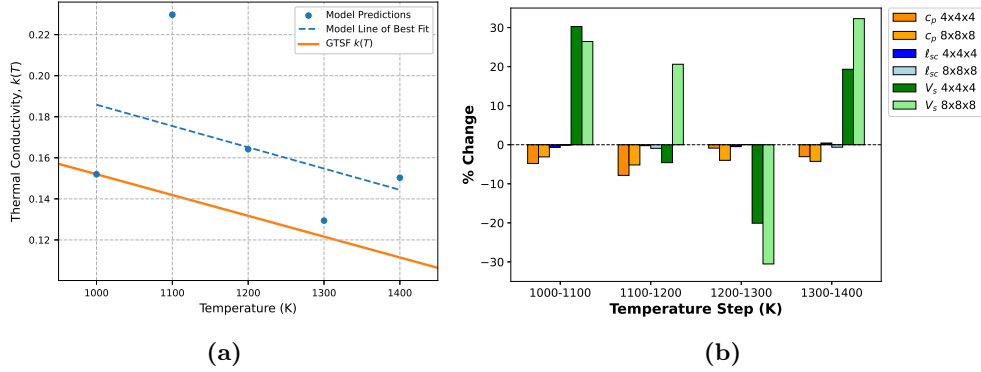


Fig. 9: (a) Results of simulating the SuperSalt “Big” salt with a $4\times 4\times 4$, 741 atom simulation space, compared to an $8\times 8\times 8$, 5933 atom simulation space, over 5 temperatures from 1000K to 1400K. There is significant variance for both predictions. (b) The percent change in each property with each temperature step, for both the $4\times 4\times 4$ and $8\times 8\times 8$ simulations spaces.

In Figure 9a, the line of best fit for $k(T)$ of individual ISM predictions is compared to the predicted slope by the GTDF, extrapolated from the hypothetical melting point at 100K intervals. In Figure 9b, the percent change between each temperature interval is shown for C_p , v_s , and ℓ_{sc} . While ℓ_{sc} minimally changes with increasing temperature, and C_p decreases at a relatively constant rate, v_s changes dramatically and without any apparent temperature dependence. In order to address this variance, the simulation was repeated with a much larger simulation space of the 8x8x8 supercell (from 741 atoms to 5933 atoms), however there was no observed change in variance of any of the properties, despite the observation of a very slight positive temperature dependence of $k(T)$ for the larger simulation.

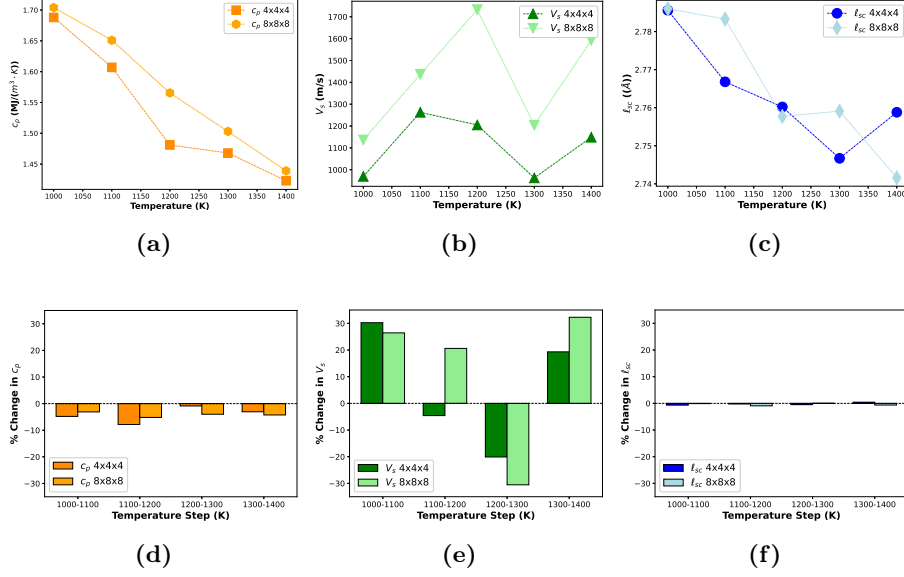


Fig. 10: “Big” SuperSalt variations in: (a) C_p at each simulated temperature, for both 4x4x4 (741 atoms) and 8x8x8 (5933 atoms) sized simulation spaces. (b) v_s at each simulated temperature, for each simulation space. (c) ℓ_{sc} at each simulated temperature, for each simulation spaces. (d) The percent change in C_p between each simulated temperature for both 4x4x4 and 8x8x8 sizes, with an average change of -4.15 and -4.13% respectively. (e) The percent change in v_s between each simulated temperature for both 4x4x4 and 8x8x8 sizes, with an average change of 6.22 and 12.2%, or average magnitude of change of 18.57 and 27.46% respectively. (f) The percent change in ℓ_{sc} between each simulated temperature for both simulation sizes, with an average change of -0.24 and -0.4%, or average magnitude of change of 0.46 and 0.43% respectively.

Between 1100 and 1200K, the change in v_s is negative for the initial 4x4x4 supercell sized space (741 atoms) and positive for the 8x8x8 supercell sized space (5933 atoms), as shown in Figure 10 where the variation in each parameter can be more clearly seen. This indicates that the variance is not a reflection of true temperature dependence but

is instead a statistical variance. It is unclear whether or not increasing the simulation size further will decrease the statistical variability, or if this represents a limit to the applicability of Bryk’s method [7] for predicting v_s . Future work will explore the effects of lengthening the simulation time and increasing the number of trials of simulation to average in determining v_s .

3.5 Temperature Dependence Results

To minimize the number of confounding factors in the ISM that can occur with salt mixtures, a unary salt was used to validate the Gheribi Temperature Dependence Function (GTDF) [30]. The GTDF shows close agreement to discrete predictions made by the ISM model at multiple temperatures. In Figure 11, the results of discrete predictions for KCl are compared to the GTDF prediction extrapolated from the predicted k_{T_m} . Although the two predictions are generally in agreement, their values are slightly higher at the melting temperature and exhibit a greater temperature dependence than the experimental correlation for KCl in the MSD-TP database [8].

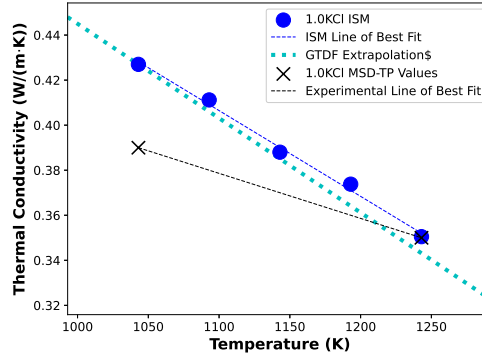


Fig. 11: Validation of the GTDF [30] for unary salts by plotting the predicted temperature dependence of the thermal conductivity of molten KCl against the predictions of the ISM model applied at 50 K, 100 K, 150 K, and 200 K above the melting temperature, and compared to experimental values from Nagasaka [64] that are reported in the MSD-TP [8].

The predictions show near agreement at 1243K, however at 1043K or (the melting temperature), they overpredict by about 9.8%, representing $0.427 \text{ W}/(\text{m} \cdot \text{K})$ compared to $0.389 \text{ W}/(\text{m} \cdot \text{K})$. Reasons for this difference are not currently understood, although it was also shown in the original SCM predictions [67] that there is a stronger temperature dependence of C_p and v_s compared to the minimal changes in ℓ_{sc} as shown in Figure 10c. It can also be observed in Figure 8 that the line of best fit for ISM predictions is noticeably different than the GTDF predicts. Future studies will need to explore the mechanisms behind temperature dependence and potential reasons for discrepancies between the GTDF, computational predictions, and experimental results.

4 Discussion

4.1 Uncertainty in Adiabatic Speed of Sound (v_s)

Adiabatic speed of sound shows the most variance and deviation from literature values among the three parameters required in the kinetic theory expression of thermal conductivity (Eq. 2). This is represented by the highest mean absolute error (MAE) compared to experimental values for NaCl-KCl among v_s , C_p , and ℓ_{sc} as can be seen in Table 3. The greatest deviation in v_s from experiment occurs in the case of MgCl_2 , where v_s is predicted to be 1003 m/s, which is 52.9% less than the 2101 m/s predicted by Gheribi as summarized in Table 2 [33]. This deviation in v_s results in significant underprediction of thermal conductivity in Mg^{2+} bearing mixtures. While the reasons for variance are clear, the large deviation from experiment are less so because there is limited availability and varying degrees of accuracy in experimental data. One additional explanation is a limited capability of the SuperSalt MLIP to represent the stiffness of the salt sufficiently for Bryk’s method to yield accurate results.

Table 3: Summary of mean absolute error (MAE), simulated time, and the parameter that presents the largest source of error in the calculation of NaCl-KCl properties using the ISM method.

Parameter	MAE	Simulated time	Largest error source
k_{T_m}	24.5%	1105 ps	v_s
$k(T)$	—	1395 ps	k_{T_m}
v_s	15.5%	540 ps	ρ
C_p	2.6 %	430 ps	ρ
ℓ_{sc}	4.1%	135 ps	pPDF

Variance of the adiabatic speed of sound predictions occurs due to difficulty associated with tracking stress fluctuations in MD simulation. Similarly to the heat current function used in the Green-Kubo method (see Supplementary Materials for additional information), measured stress fluctuations are highly vulnerable to statistical variance, leading to convergence at different values between trials [22]. While the longitudinal stresses associated with Bryk’s stress fluctuation method are still easier to track than the heat current [15] used in Green-Kubo, the resulting variance is significant enough that multiple trials are required to reach comparable accuracy to experimental measurements. For most salts, three trials or sets of simulations with a 3x3x3 supercell is sufficient to reach a converged value. However, the variance shown in Figures 6b and 9b suggests that additional trials or other improvements may be needed for Mg-bearing or extremely disordered mixtures. Future work will explore optimization of this part of the workflow.

The speed of sound in most alkali halides were measured by Bockris et al. in 1957 [6], with the exception of MgCl_2 . To the best knowledge of the authors, the only experimentally measured value for MgCl_2 is from Sternberg et al. [83], measured in 1969. When plotted together according to group (I or II) and period (see Figure 12),

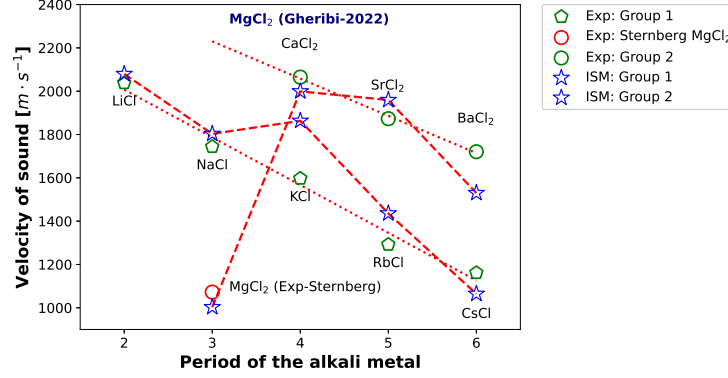


Fig. 12: The measured adiabatic speed of sound for group I and II chlorides, plotted by period. While the trend for group II could indicate that MgCl_2 should have a higher v_s than experimental measurements show, ISM predictions show agreement with all experimental measurements, including the abnormally low MgCl_2 value

the measured v_s of alkali halides form seemingly linear trends. The exception is MgCl_2 , which is less than half of the expected value assumed by the trend.

When paired with certain experimental thermal conductivity measurements for MgCl_2 , which cluster around $0.5 \text{ W}/(\text{m} \cdot \text{K})$, it appears plausible that the v_s of MgCl_2 should be higher. When paired with c_p and ℓ_{sc} as calculated by the ISM ($1.69 \text{ MJ}/(\text{m}^3 \cdot \text{K})$ and $4.54 \text{ \AA}$, respectively), a v_s of 1048 m/s results in a prediction of $k = 0.26 \text{ W}/(\text{m} \cdot \text{K})$, a 46% underestimation.

Following similar reasoning, Gheribi proposed that the experimental value by Sternberg is unreliable and should not be used in predictive models [29]. By running an EMD simulation using a Birch–Murnaghan equation of state, Gheribi derived a predicted v_s of 2101 m/s , which fits much more closely to the trend shown in Figure 12. This higher value was utilized in the initial predictions of the SCM [67]. While this higher value results in predictions closer to certain experimental values, not all experimental values for MgCl_2 are close to $0.5 \text{ W}/(\text{m} \cdot \text{K})$, such as $1.018 \text{ W}/(\text{m} \cdot \text{K})$ by Filatov [26] and $0.198 \text{ W}/(\text{m} \cdot \text{K})$ by Golyshev [31]. Further experimental measurements are necessary to reduce the uncertainty of the true k_{T_m} of MgCl_2 .

The ISM consistently predicts a v_s near 1000 m/s , which is much closer to Sternberg’s experimental measurement. To validate this prediction, the simulation time was extended from 150 ps in the NVE ensemble to 1000 ps , and the space increased from a $3 \times 3 \times 3$ to $7 \times 7 \times 7$ supercell (324 atoms to 4116 atoms). However, the predicted values did not change. This indicates that if the SuperSalt MLIP can be assumed to accurately represent the stiffness of MgCl_2 , the true v_s may be closer to the experimental measurement by Sternberg. The predictions for other alkali halides by SuperSalt are close to the experimental measurements by Bockris (Figure 12), which strengthens the case for SuperSalt being sufficiently trained to represent MgCl_2 accurately.

Recent developments in understanding of the structure of MgCl_2 illuminate potential reasons for the seemingly abnormally low v_s predicted by the ISM and measured

experimentally. It is now well documented that MgCl_2 and mixtures containing Mg^{2+} such as MgCl_2 -KCl exhibit intermediate range order [73, 76, 93, 96]. Roy et al. confirmed that MgCl_2 forms distorted square pyramidal polyhedra, with a preferred coordination number of 5 [69], and attributes this complex local structure as responsible for certain experimental studies that appear contradictory. Gheribi’s equation of state based EMD prediction of v_s was performed with a simulation size of 144 atoms and over time scales of 30 ps per volume step. This size and time scale may be too small to allow for sufficient relaxation of the complexes formed by Mg^{2+} , which would result in artificially high v_s predictions. It seems plausible that a low v_s of MgCl_2 is a reflection of unique intermediate order rather than failings of experimental measurement or the capability of the SuperSalt MLIP to represent the structure of MgCl_2 and Mg^{2+} bearing mixtures.

4.2 Potential Limitations of the ISM

When the thermal conductivity predictions of the ISM model are compared to experimental values across varied composition of NaCl-KCl in Figure 5, it is interesting to note close agreement at each endmember, but increasing disagreement as the complexity of the salt increases. While the ISM model predicts a minimum at the NaCl-KCl eutectic point, experimental values as well as RNEMD show a more linear relationship between 0 and 100 % molar NaCl [64, 72].

RNEMD was performed with the SuperSalt potential in order to compare the ISM model to existing predictive methods. Due to limitations in available memory, the largest simulation size utilized was 3,456 atoms. This likely led to phonon truncation and thus underestimation of the thermal conductivity, as documented in literature [10, 78, 82]. Despite the underestimation in scale, the correlation with increasing molar concentration of NaCl agrees with both experimental and other RNEMD values from Ding et al. and Li et al. [20, 50]. Because the RNEMD was performed with the same SuperSalt potential as the ISM model, these results point to potential refinement in the way the SCM approximates the change in mean free path of the energy carrier as the composition of the mixture changes.

As seen in Figure 4a, the greatest discrepancy between experimental or RNEMD values with the ISM model’s predictions is between the 0.3NaCl-0.7KCl and 0.7NaCl-0.3KCl compositions. In Figure 4b, while C_p and v_s do not decrease consistently, ℓ_{sc} decreases significantly, reaching a minimum at 0.5NaCl-0.5KCl. The extent of this minimum appears to be an exaggerated decrease in ℓ_{sc} , assuming that the actual thermal conductivity of 0.5NaCl-0.5KCl is within the estimated experimental uncertainty shown.

The driving force for the decrease with compositional mixing is the phonon transfer decoherence factor b_{PH} . The current model assumes that if a phonon-like propagation encounters a cation of a different species, the phonon is completely disrupted due to differing phonon harmonic oscillator frequencies. This approximation was originally made due to the difficulty in quantifying the degree to which vibrational transfer is compatible across differing cation-anion domains. In order to simplify quantification, it was left in a binary state of either complete or no disruption. While this method predicts within 5-10% accuracy for the actinide bearing salt NaCl- UCl_3 , 2.4% for FLiNaK,

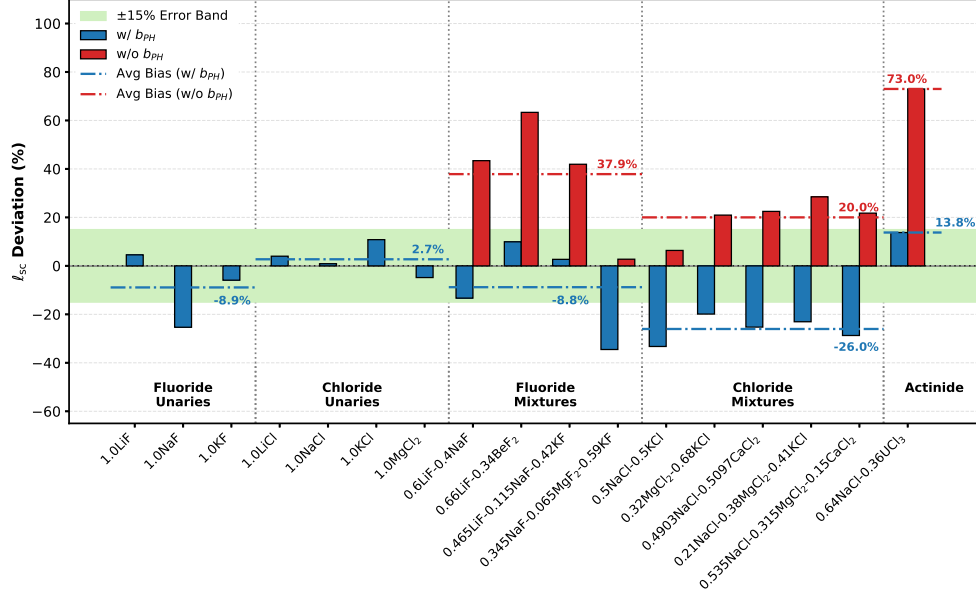


Fig. 13: Percent deviation of the structural coherence length ℓ_{sc} compared to experimentally calculated values for different types of halide salts. Exclusion of the b_{PH} factor generally results in an overestimation, while inclusion of the factor exhibits both over- and underestimations.

and 7.8% for FLiBe, the results in the SCM paper show a consistent underestimation of between 25-35% across binary and ternary mixtures such as 0.673LiCl-0.367KCl, 0.5NaCl-0.5KCl, 0.49NaCl-0.51CaCl₂, and 0.535NaCl-0.315MgCl₂-0.15CaCl₂ [67].

A potential explanation for the uneven applicability of b_{PH} is that for actinide-bearing salts or fluorides, the difference in phonon harmonic oscillator frequency between cation species is large enough that the current binary approximation sufficiently captures the degree of phonon transfer. However, in the case of mixtures with more a similar phonon frequency or atomic mass, the binary quantification of phonon transfer may not be as accurate. To improve applicability, b_{PH} may need to be quantified differently to account for the non-zero possibility that phonons can transfer significantly across cation-anion chains comprised of differing cation species. This may occur if their harmonic oscillator frequency is similar and thus results in structural coherence of the energy carrier.

If the phonon transfer factor is set to 0 (i.e. it is assumed that phonon transfer is not impacted by different cation species), the ISM model's thermal conductivity predictions match experimental and BHMTF-based RNEMD values more closely in the case of alkali chlorides, as can be seen in Figure 13. This can also be seen as functions of composition in Figure 4a and temperature in Figure 8. Assuming that the experimental and RNEMD values are correct, further development of the b_{PH}

factor could potentially improve the SCM’s performance as the ℓ_{sc} is more accurately approximated.

5 Conclusions

The proposed Integrated Structural coherence and Machine learning interatomic potential (ISM) model serves as a proof of concept for integration between the structural coherence model (SCM) and a semi-universal Machine Learning Interatomic Potential (MLIP). By using the SuperSalt MLIP to generate MD-derived structural and thermophysical inputs, the ISM workflow expands SCM predictions beyond the limited set of compositions with existing experimental or computational property data. This enables systematic investigation of thermal conductivity across variations in cations, composition, and temperature. The integrated model demonstrates that by predicting density, heat capacity, adiabatic speed of sound, and the mean free path informed by a partial pair distribution function, thermal conductivity can be predicted more quickly and with accuracy comparable to current experimental and MD methods.

To expand this predictive capability and further reduce computational costs, we validated Gheribi’s temperature dependent function (GTDF). To do this, isothermal bulk modulus was simulated and paired with other properties to approximate the Grüneisen parameter, allowing the extrapolation of predicted thermal conductivity at the melting temperature to additional temperatures. While showing close agreement to ISM predictions at discrete temperatures for KCl, both ISM and GTDF prediction disagreed with experimental temperature dependence. Future work will be necessary to explore potential reasons for this disagreement.

The ISM model required 1/20 the computational cost of an RNEMD simulation performed with the same SuperSalt potential, demonstrating the degree to which computational cost is reduced by using this method. The predictions for most salts align closely with the existing SCM, with notable increased variance and deviation in the predictions of v_s . However, this variance in v_s obfuscates the trend of model’s predictions of thermal conductivity with respect to both temperature and composition. Future development will focus on improving predictions for the v_s of magnesium-bearing and highly complex salts. Additional development will also need to be conducted to better quantify the b_{PH} decoherence factor. This is because the current implementation of the factor appears to unevenly quantify the degree to which compatible phonon transfer is present for simple chloride mixtures compared to fluorides and actinide bearing salts. Future application of the model ISM to MLIPs trained specifically for actinide bearing and fluoride salts is also planned, which will facilitate this development.

Uncertainty in both experimental measurements and computational predictions of thermal conductivity and adiabatic speed of sound prevents clear conclusions from being drawn on molten salt’s thermophysical properties. In order to verify and improve current model predictions, targeted experimental and computational studies need to be conducted to address these uncertainties. Of particular value would be the establishment of a standardized methodology for molten salt thermal conductivity measurements.

Once validated, the proposed ISM model demonstrates potential for populating current thermophysical property databases and improving understanding of molten salt thermal conductivity. As thermal conductivity is more deeply understood, energy generation systems are able to be designed with more highly optimized safety and efficiency.

CRediT authorship contribution statement

Isaac Walker: Conceptualization, Methodology, Software, Formal Analysis, Investigation, Writing – Original Draft, Visualization. **Jacob Numbers:** Conceptualization, Writing – review and editing. **Nathan Burlett:** Software, Formal Analysis, Investigation, Writing – review and editing. **Anthony Birri:** Validation, Writing – review and editing. **Troy Munro:** Supervision, Conceptualization, Validation, Funding acquisition, Writing – review and editing.

Author Declarations

During the preparation of this work, the authors used Gemini to search for sources, synthesize literature, identify articles of interest, assist with conceptual research, support code development, and format content. After using these services, the authors reviewed and edited the content as needed and take full responsibility for the content of the published article.

The authors have no conflicts of interest to disclose.

6 Data Availability

All code needed run the ISM model through a HPC cluster is publicly available at the repository: <https://github.com/BYU-TEMP-Lab/2026-ISM-Integrated-SCM-MLIP>. For questions regarding set up of the model contact Isaac Walker at ihwalker@byu.edu.

Acknowledgments

Funding for this research has come from the Department of Energy, Nuclear Energy University Program (NEUP): Award #DE-NE0009502. Jacob Numbers and Anthony Birri are supported by the Molten Salt Reactor Program and the Nuclear Energy Advanced Modeling and Simulation Program under the US Department of Energy's Office of Nuclear Energy.

Meaningful discussions with Matthew Emerson and Dane Morgan provided valuable direction for the project.

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