

Simultaneous Prediction of Density, Viscosity and Heat Capacity of Ionic  
Liquids- A Deep Learning Approach

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Abstract

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Estimation of properties of ionic liquids with artificial neural networks have been successful in overcoming the challenges posed by experimental predictions and equation of state models. With the advent of deep learning and affordable GPU-computing, there is potential to accelerate the predictions with better accuracy. In this work, different deep neural network architectures have been designed to determine the density, viscosity and heat capacity of ionic liquids (ILs) as single task models in the Python package Keras. The features for the ILs include temperature, pressure and molecular descriptors of the cation and anion. Additionally, multi-task neural networks were designed to simultaneously predict all three properties. The root mean squared error and R-squared have been used to evaluate the various models. The performance of the best multi-task model is compared to the best single task model for each property. Viscosity, heat capacity perform better in single task and density performs better in multi-task. Overall, multi-task learning shows to be promising and can be further improved by including more properties.

## 1. Introduction

Ionic liquids (ILs) are organic or inorganic, paired ions that are liquid at or above room temperature. They are popularly called 'green solvents' or 'solvents of the future' due to their exceptional properties; negligible vapor pressure, high chemical stability and wide liquid temperature range.[1][2]

To evaluate the ILs utility in applications, various physicochemical properties need to be evaluated. To do that experimentally is expensive because the combinatorial space of anions and cations can be very large.[3] Particularly density, viscosity and heat capacity are important properties of liquids for industrial design and operation and need to be evaluated before the liquids can be employed in processes.[2][4][5] Moreover, these properties are temperature and pressure dependent and gathering experimental data for the possible combinations is an insurmountable task. To accelerate the property predictions for ILs, various equation of state models began to be evaluated. [6] These models were highly accurate but could not be generalized across different ILs. Hence mathematical models and the use of artificial neural networks have been employed to tackle these issues.

Most of the work using artificial neural networks for IL property prediction in literature has been done for predicting a single property of ionic liquids. Traditionally, artificial neural networks with or without group contribution

methods have been used to evaluate properties of ionic liquids.[1][7] However, there has been limited success in building a single robust model that can be successful in predicting various properties of the wide variety of ionic liquids (multi task) . The term 'multi-task' indicates that the model can predict multiple outputs for tasks that are related to each other through their feature space. The advantage of multi-task modelling is that it optimizes time and computing power by having to train 1 neural network as opposed to multiple single task networks. Kianfer et al[8] have demonstrated the use of artificial neural networks to predict density, viscosity and electrical conductivity of pyridinium based hydrophobic ionic liquids. The authors train on 36 ILs and predict on 12 ILs with only temperature as input to give R-squared values of  $\sim 0.99-1$  for all properties. Moreover, Cancilla et al[9] have developed what they call a 'global model' for amino acid based ionic liquids with 112 samples. The input to the global model is temperature, anion polarizability and cation polarizability which predicts viscosity, density and thermal expansion coefficient. However, Gonzalo et al[10] have done a multitask prediction of density, viscosity and refractive index which showed a decrease in predictive power compared to their single task predictions and recommend the use of different neural networks for each task. This could be held true before the recent advent and

popularity of deep learning which is making multi task learning more powerful in some applications.[11][12]

A deep neural network is a multi-layer artificial neural network. It has been extremely successful in image and speech recognition and is considered the state-of-the-art modeling technique in machine learning. [13] Its application in the field of QSAR (Quantitative Structure Activity Relation) and QSPR (Quantitative Structure Property Relation) predictions for drug discovery have been successful. [14][15] Due to open source databases, accelerated GPU computing and optimization algorithms available with open source packages, deep learning has become easier to extend into domain sciences and replace machine learning algorithms with better accuracy. [12]

In this work, we have demonstrated a multi-task feed forward deep neural network to evaluate the predictions of heat capacity, density and viscosity of ionic liquids. Having a network that can predict multiple properties for ionic liquids is beneficial for exploring various types of ionic liquids at different temperature and pressure. This network also leverages the representation learning ability of deep neural networks and hence circumvents the need to calculate interaction energies or interaction terms through other computational techniques.[16][17] Neural networks are powerful because they have the ability to learn and make intelligent

decisions on their own and can be re-designed till accuracy has reached the optimum value.

In this work, we have explored the simple MLP ,2 versions of merged architectures and have explained them as: 1) The simple multi-layer perceptron 2) The first merged architecture (ILnet1) 3) The second merged architecture (ILnet2).

For each of the 3 architectures, we performed the single task predictions of heat capacity, density and viscosity. The performance of the various architectures is calculated and compared using the root mean squared error (RMSE) and R-squared. Ultimately, the best network architecture is selected based on the prediction of the single task models and used as the architecture for multi-task modelling. Further single and multi-task models are compared for their accuracy across each property.

## 2. Methods

### 2.1 Data collection

The open source ILThermo database is a web based ionic liquids database which is provided by the National Institute of Standards and Technology (NIST).[18] The data was collected by web scraping for each property as a separate dataset. However, this dataset is the overlap of the 3 datasets. It has ~23,000 ILs at different temperature, pressure, heat capacity, viscosity and density values. 25% of these ILs were reserved as a test dataset and the remaining was used in 5-fold cross validation. The temperature ranges from 278.15-373.15K and pressure ranges from 100-20000 kPa. The heat capacity, density and viscosity values range from 231.8-1764 J/k/mole, 847.5- 1557.1 kg/m<sup>3</sup> and 0.00316- 10.2 Pa-s respectively.

The features or molecular descriptors for the cations and anions were generated from the open source package RDKit.[19] Molecular descriptors are calculated features of a molecule that encapsulate its activity and molecular structure. They are key in *insilico* drug discovery and QSAR modelling. [20][21] All the features were scaled to have a mean of zero and unit variance. This prevents any one feature from dominating the feature space. Further, the output data of heat capacity, density

and viscosity data were log transformed because the data was not normally distributed.

## 2.2 Feature /Data generation

Salty[22]-an interactive data exploration tool for ionic liquid data from ILThermo(NIST)[18] which is built on top of RDKit[19], the popular cheminformatics software has been used to generate the features. Using Salty, we generated 94 descriptors for the cation and anion respectively as shown in Table 1.

Feature Group	Number of Descriptors
Topology	3
Kappa	4
Connectivity	12
Constitutional	11
MOE-type	58
Molecular Property	2
Other	4

Table 1. Highlights the distribution of the features used for modelling

## **2.3 Network Architecture and Design**

Network architecture plays a key role in determining the accuracy of neural networks and various hyper parameters need to be tuned before a network can predict accurately. The networks were built in Keras[23] which is a deep learning API written in Python. Figure 1 illustrates the different architectures explored.

### **2.3.1 Simple Multi-Layer Perceptron**

This network architecture (Figure 1.a.) consists of a feed forward MLP taking cation descriptors, anion descriptors and state variables of temperature and pressure as input data. For the MLP architecture, a grid search was performed of (2,3,4,5) fully-connected layers, and (16,32,64,128,256,512) neurons per layer with ReLU activation functions. To initialize the weights, glorot normal was selected in each of the dense layers. To prevent over fitting, a dropout of 0.5 was added after each fully connected layer and an early stopping criterion was incorporated. Ultimately the best model which had the lowest validation error rate was selected.

### **2.3.2 ILNet1 architecture**

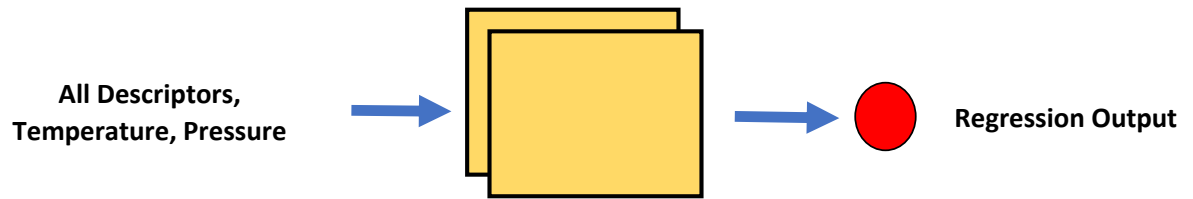
This architecture (Figure 1.b.) consists of 2 separate MLP networks of the best architecture selected from the above grid search. These 2 networks take cation

descriptors and anion descriptors, respectively, as input. The state variables of temperature and pressure are then concatenated with the outputs of those 2 networks before being parsed into a regression neuron with a linear activation function in the last layer. Similar initialization, dropout and activation was employed as the Simple MLP for each layer.

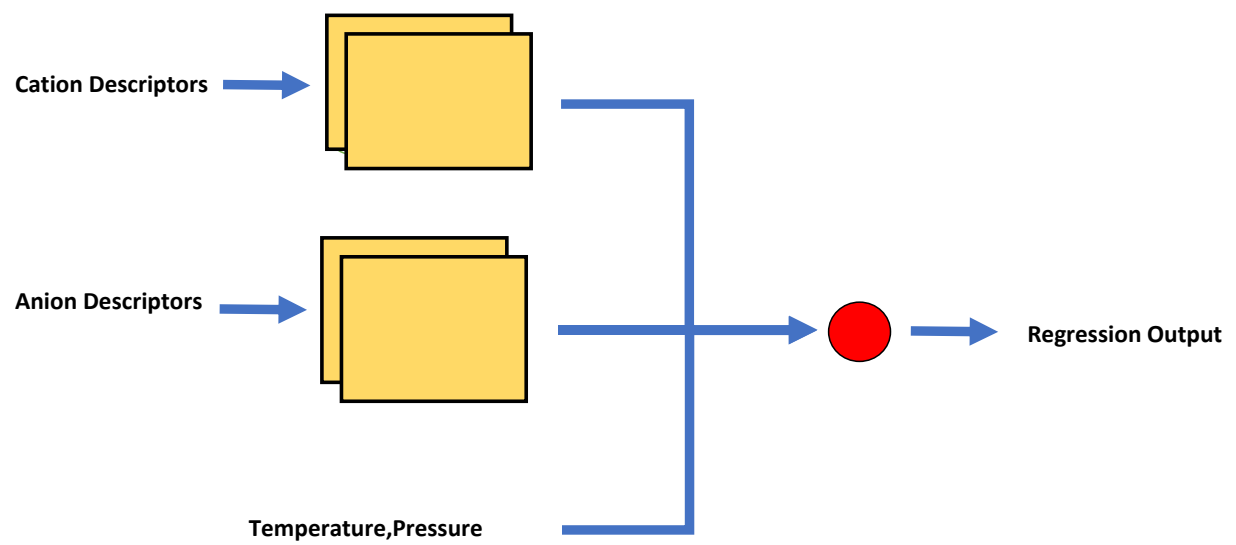
### **2.3.3 ILNet2 architecture**

This architecture is like the Merged 1 architecture but before the final regression neuron, there are 2 additional fully connected layers (Figure 1.c.). This allows the combined input from the previous networks to leverage the representation learning ability of deep neural nets and develop their own interaction features between the cation descriptors, anion descriptors and state variables in these fully connected layers.

1.a.



1.b.



1.c.

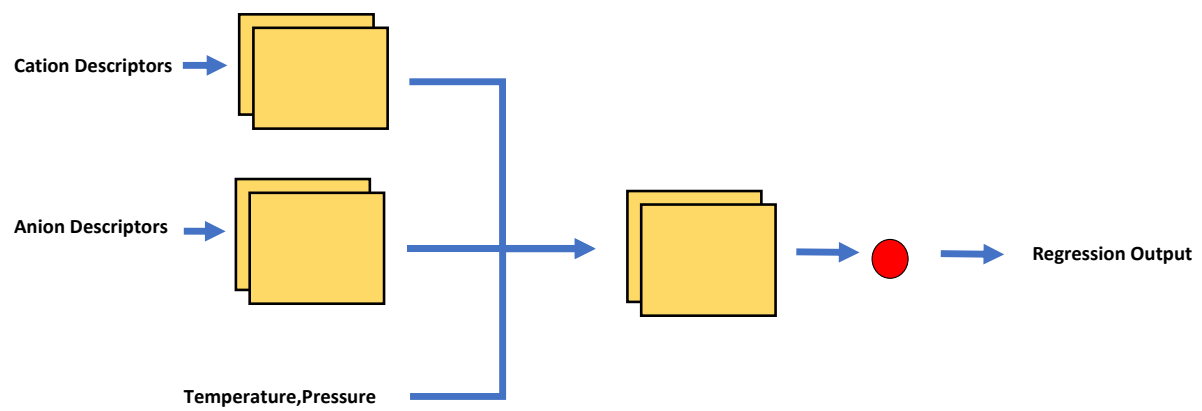


Figure 1. shows the a. MLP, b. ILNet1 and c. ILNet2 architectures respectively

## 2.4 Training protocol

The network was created using the Keras 1.2 functional API interface[23]. From the original dataset, 25% were split out as testing data and the remaining 75% were used for 5-fold cross validation. During the 5-fold cross-validation, an early stopping criterion was evaluated: if the validation loss did not improve for 50 epochs, the weights of the last best model was saved as the final model. The 'Adam' optimizer was used to train the network for 500 epochs with the default settings and in a batch size of 30. It was trained using a Tensorflow backend with GPU acceleration using NVIDIA CuDNN libraries.

## 2.5 Loss function and Regression Metrics

A mean squared error loss function was employed while training the models. However, the RMSE and R-squared have been used as the metrics to compare the performance between different architectures as well as compare the performance of individual properties. The formulae for the following are:

$$\text{RMSE} = \sqrt{\frac{\sum (y_{pred} - y_{exp})^2}{N}} \quad \text{R-squared} = 1 - \frac{\sum (y_{exp} - y_{pred})^2}{\sum (y_{exp} - y_{mean})^2}$$

### **3 Results and Discussion**

#### **3.1 Model Architectures and Hyper Parameter Tuning**

Prediction accuracy highly depends on the network architecture. Figures 2-4 show the performance of the 3 architectures with RMSE and R-squared for single task predictions of heat capacity, density and viscosity respectively. Each of the metrics reported have been recorded for the hyper parameters that best optimized its performance and accuracy. Table 2 shows the various neurons per layer for the single networks. For the single MLP, 2 dense hidden layers were recorded as the optimum number for each property. The initialization, activation and dropout were all the same as mentioned in 2.3.1. In the last layer, 1 neuron with linear activation was employed. For the ILnet1 architecture, each of the networks to process the cation and anion descriptors had 2 dense hidden layers. Further, for the ILnet2 architecture, there were 2 fully connected layers with a dropout of 0.1 after each layer before the final layer, the rest being the same as the ILnet1. In Table 2. the numbers indicated for ILNet2 show the neurons per layer for processing the cation/anion networks (before the hyphen) and the fully connected layers after they are merged (after the hyphen) respectively.

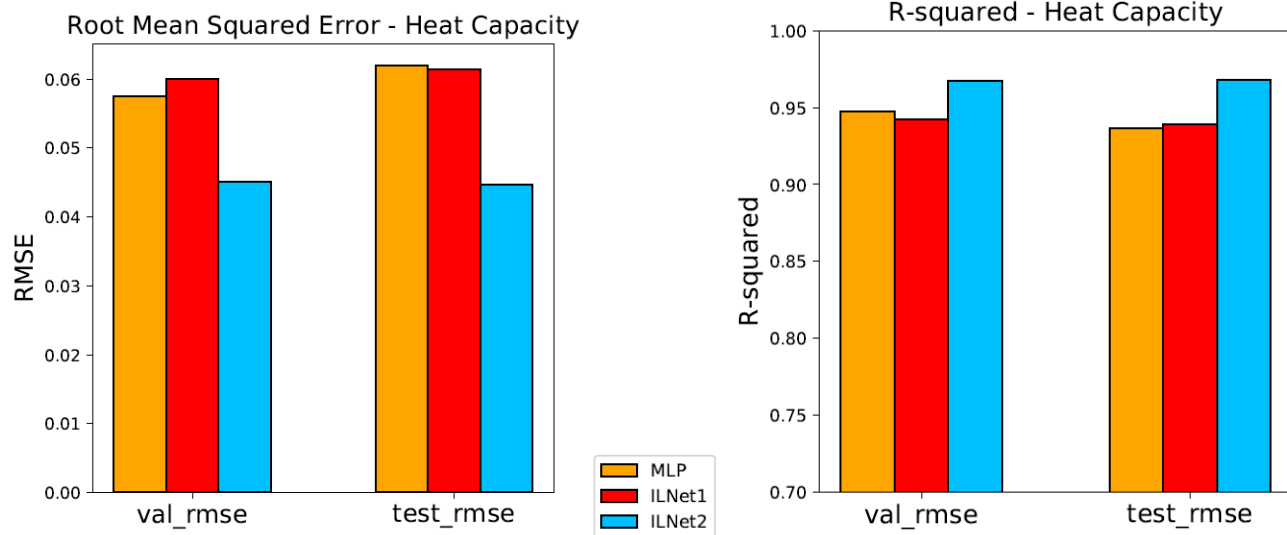


Figure 2. shows the RMSE and R-squared for MLP, ILNet1 and ILNet2 for Heat Capacity predictions

	MLP	ILNet1	ILNet2
Heat Capacity	64	64	64-64
Density	64	128	128-64
Viscosity	128	64	64-128

Table 2. shows the number of neurons per layer for the most optimized networks in the grid search for MLP, ILNet1 and ILNet2.

### 3.2 Single Task Predictions

The 2 metrics evaluated to compare the regressions have been explained in 2.5 and shown in Figures 2-4 for heat capacity, density and viscosity respectively. In Figure

3 it is observed that the MLP and ILNet2 architectures have similar performance on the density test set. However, ILnet2 architecture has the best performance on the heat capacity and viscosity test sets as seen from Figures 2 and 4.

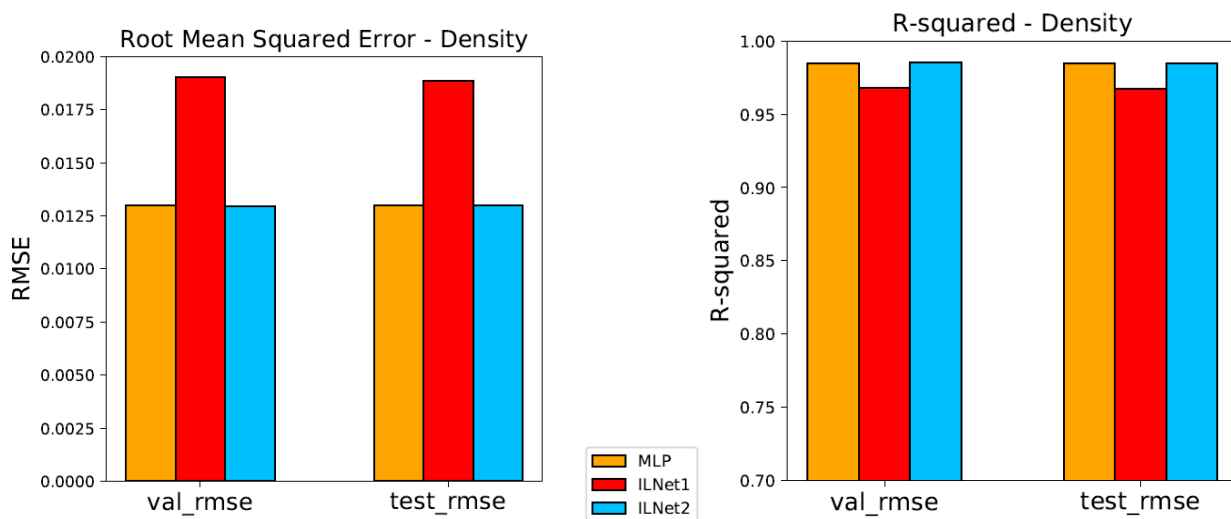


Figure 3 shows the RMSE and R-squared for MLP, ILNet1 and ILNet2 for Density predictions

This indicates that ILNet2 is performing well across all 3 properties and MLP performs well for density too. Hence evaluating both the MLP and ILNet2 architectures further for multi-task predictions was necessary.

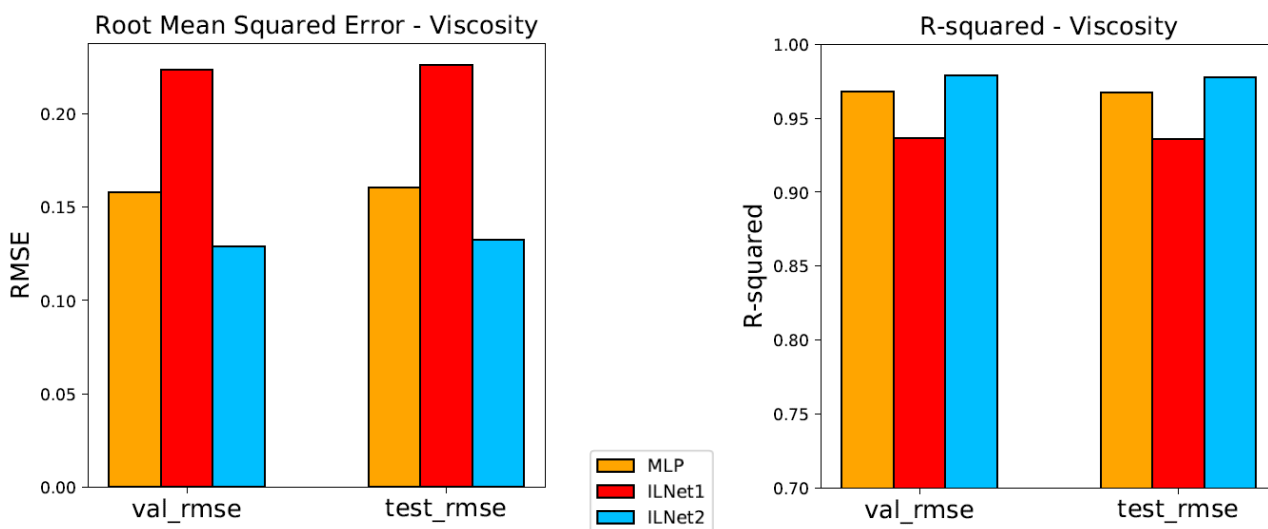


Figure 4 shows the RMSE and R-squared for MLP, ILNet1 and ILNet2 for Viscosity predictions

### 3.3 Multi-task Predictions

Multi-task predictions are done for tasks that are related to each other through their feature space. It is the ability of neural network architectures to exploit commonalities and differences between related tasks. To extend the analysis to multi-task, the MLP and ILnet2 architectures were used with the only difference that the networks were modified to have 3 outputs for each of the 3 tasks. The models were then tuned and optimized to achieve accuracy. The optimized hyper parameters for ILNet2 were 128 neurons per layer for the dense layers that process the cations and anions and 128 neurons per layer for the fully connected layers before the 3 outputs. For the MLP, it was simply 128 neurons per layer for the dense

layers. Figure 5 compares the performance of MLP and ILNet2 architectures across all 3 properties. It can be seen that ILNet2 outperforms MLP for both RMSE and R-squared. This indicates that ILNet2 has been most accurate in not only doing multi-task task predictions, but it can also be attributed to the fact that the fully connected layers in the end enhanced its learning ability.

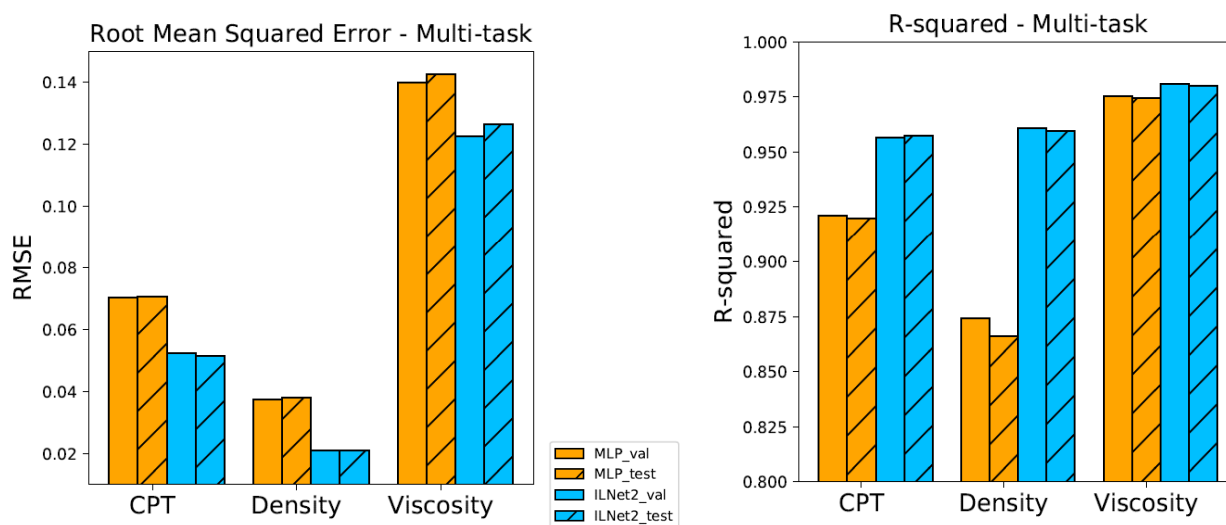


Figure 5 shows the performance of MLP and ILNet2 architectures for each of the 3 properties

### 3.4 Single v/s Multi-task Predictions

From a modelling perspective, having to generate different hyperparameters for single tasks can be time consuming and compute intensive as opposed to when it's done for multiple tasks at once. While maintaining that this is an advantage, it is

important to compare the property predictions between single and multi-task. The relative change in the RMSE with respect to the single task predictions has been shown.

$$\% \text{ Relative Change} = \frac{(RMSE_{single} - RMSE_{multi}) * 100}{RMSE_{single}}$$

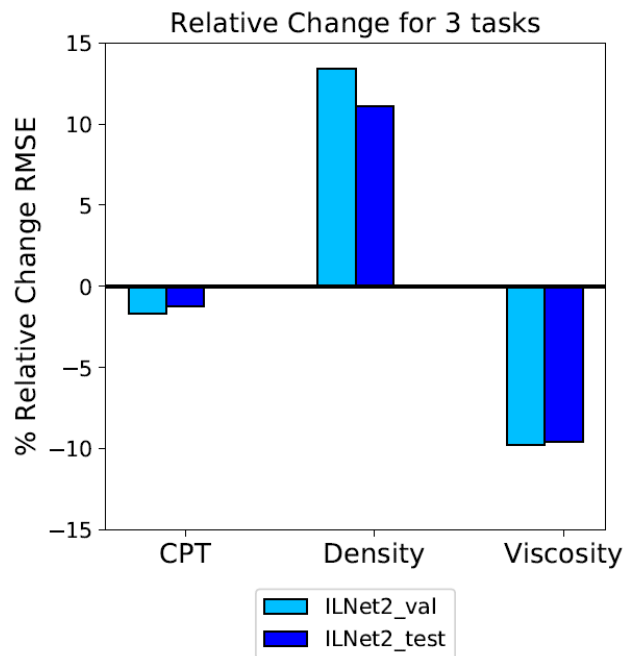


Figure 6 shows that density has a positive relative change and viscosity and heat capacity have a negative relative change.

A positive relative change indicates that multi-task has been more accurate in predicting that property. Here density has been better predicted with multi-task. A negative relative change indicates that single task has been more accurate in predicting the property, which is seen in heat capacity and viscosity.

It is commonly known that, multi task prediction is based on the idea that related tasks can improve performance by inductive bias.[11] It is also shown to outperform single tasks in some applications.[15] There is also evidence of multi-task predictions not being able to outperform single task.[10][24] Most of the conclusions heavily depend on the data being used. The reason multi task is not outperforming single tasks by a large margin is because the experimental values are gathered from many sources and hence learning inherent task relatedness can be difficult. ILThermo[18] has combined multiple experimental values from various authors and that has introduced what is commonly known as the irreducible error in these models. Another important fact to highlight is that deep learning algorithms are shown to improve with larger datasets. Data sets with millions are used for evaluating multi-task learning. However, accurately labelled data is usually difficult to obtain in such early stage research initiatives.

## 4 Conclusion

We have shown multi task and single task predictions for heat capacity, density and viscosity and evaluated the models' performances. Multi task is best when exploring a wide variety of properties and can be easily extended by adding other properties. Depending on the accuracy requirements, both single and multi-task show promising results. Evaluating several properties of ionic liquids is imperative if we want to employ these solvents and design more of them for clean energy applications. In order to accelerate the development process, determining these properties from multi-task deep neural networks can be a cost effective, viable and a long-term solution to enhance experimental research. Further, as more data becomes available, the models can be retrained to give better accuracy and expand its range of IL predictions.

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