# Multi-Scale Neural Model for Tool-Narayanaswamy-Moynihan Model Parameter Extraction

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**Abstract.** Glass transitions are an important phenomenon in amorphous materials with potential for various applications. The Tool-Narayanaswamy-Moynihan (TNM) model is a widely used empirical model that describes the enthalpy relaxation behavior of these materials. However, determining the appropriate values for its parameters can be challenging. To address this issue, a multi-scale convolutional neural model is proposed that can accurately predict the TNM parameters directly from the set of differential scanning calorimetry curves, experimentally measured using the sample of the considered amorphous material. The resulting Mean Absolute Error of the model over the test set is found to be 0.0252, indicating a high level of accuracy. Overall, the proposed neural model has the potential to become a valuable tool for practical application of the TNM model in the glass industry and related fields.

**Keywords:** Tool-Narayanaswamy-Moynihan (TNM) model, multi-scale neural model, theoretical kinetic, enthalpy relaxation dynamics, glass transition, differential scanning calorimetry (DSC) data, deep learning

## 1 Introduction

Glass transitions are a key phenomenon in the field of amorphous materials, which have significant potential for use in a wide range of applications, including electronics, medicine, and industry [4, 11]. These materials lack long-range order, and as a result, exhibit unique properties that make them attractive for various applications. One of the most important and defining properties of amorphous materials is their glass transition temperature, below which the material behaves as a glassy solid, and above which it behaves as a supercooled liquid.

The glass transition temperature is a key parameter that influences the mechanical, thermal, and transport properties of amorphous materials. As such, precise characterization of glass transition temperatures and their associated parameters is essential for understanding and utilizing these materials.

The Tool-Narayanaswamy-Moynihan (TNM) model [5, 6] is a widely used empirical model that describes the enthalpy relaxation behavior of amorphous materials in the vicinity of their glass transition temperature. The TNM model is based on the concept of structural relaxation, which occurs when the material transitions from a metastable state to a more stable state. During structural relaxation, the enthalpy of the material decreases as it approaches a state of thermodynamic equilibrium.

Despite its widespread use, the TNM model suffers from a number of limitations. One of the main challenges in using the TNM model is determining the appropriate values for its parameters. This task is often complicated by the fact that the parameters are interdependent, and may vary depending on the material and the experimental conditions. As a result, precise determination of TNM model parameters is essential for accurate predictions of enthalpy relaxation behavior and glass transition temperatures.

One method for determining the TNM parameters is the enthalpic differential scanning calorimetry (DSC) [3], which measures the heat absorbed or released by a material as it is subjected to a controlled temperature program. By repeatedly subjecting the material to temperature cycles and measuring the enthalpy response, one can obtain the differential scanning calorimetry curve, which can then be analyzed to extract the TNM parameters. However, this process is time-consuming and requires skilled personnel to obtain accurate and reproducible results.

To address this challenge, a sophisticated artificial neural network can be developed that is trained to predict the TNM parameters directly from the set of DSC curves. This approach has the potential to significantly reduce the time and labor required for TNM parameter determination, while also improving the accuracy and reproducibility of the results.

In this article, we propose the idea to implement a neural network model, more precisely, a multi-scale convolutional neural model [1], that can accurately predict the TNM parameters of glass-forming materials from the set of their DSC curves. We believe that this model has potential to become a valuable tool for the practical application of the TNM model in the glass industry and other related fields.

## 2 Matherials and methods

### 2.1 TNM model and its parameters

The TNM model can be expressed by the following equations.

$$\Phi(t) = \exp\left[-\left(\int_0^t \frac{\mathrm{d}t}{\tau(T, T_f)}\right)^{\beta}\right], \tag{1}$$

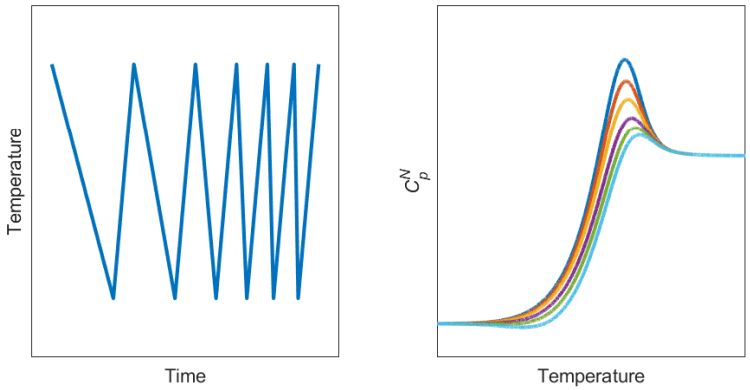


**Fig. 1.** Temperature history (left) and corresponding set of DSC curves (right) used for studying relaxation behaviour of amorphous materials.

$$\tau(T, T_f) = A \cdot \exp\left[\chi \frac{\Delta h^*}{RT} + (1-\chi)\frac{\Delta h^*}{RT_f}\right], \tag{2}$$

where: $\Phi(t)$ is the relaxation function of the given property, $t$ is time, $\tau$ is the relaxation time, $\beta$ is the nonexponentiality parameter, $A$ is the pre-exponential factor, $\chi$ is the nonlinearity parameter, $\Delta h^*$ is the apparent activation energy of the structural relaxation, $R$ is the universal gas constant, $T$ is temperature, and $T_f$ is the fictive temperature, which is defined as the temperature of the undercooled liquid with the same structure as that of the relaxing glass.

Each phenomenon described by the TNM model is determined by four parameters: $A$, $\Delta h^*$, $\chi$ and $\beta$. The other variables of the model are either determined by the ambient conditions or are universal natural constants. See [8] for the detailed information about this model and its features.

The parameters of the TNM model can be determined either by direct curve-fitting or by using some indirect non-fitting methods. A list of the most commonly used methods is discussed in [8]. It should be noted that all known classical methods suffer from some shortcomings, such as being computationally expensive, providing insufficient accuracy of model parameters, or providing only one or two of the required four parameters.

In order to get the set of TNM model parameters, it is always necessary to experimentally obtain suitable data which sufficiently describe the behaviour of the amorphous material. One of the most often used methods for the data acquisition is an application of the so-called classic cycles, where a sample of the amorphous material is repeatedly cooled at various cooling rates, while the heating scans are always performed at the same rate (see left part of Fig. 1). Then, the normalized heat capacity $C_\mathrm{p}^\mathrm{N}$ against the temperature during each cooling and heating step is observed. This response is called the DSC curve. A general example of the empiric results is shown in Fig. 1. Note that the $C_\mathrm{p}^\mathrm{N}$ curve

is slightly different for each temperature run, i.e., the material shows a memory effect.

The aim of this study is to provide a multi-scale convolutional neural model, which will process the DSC curves (right part of Fig. 1), and, consequently, it will provide four parameters of interest: $A$, $\Delta h^*$, $\chi$ and $\beta$.

### 2.2 Multi-scale convolutional neural model

Multi-scale neural networks have recently emerged as a promising approach for signal processing tasks that involve data with complex, multi-scale structures.

One of the key advantages of multi-scale networks is their ability to handle signals with varying frequency content. By using multiple layers of processing that operate at different scales, multi-scale networks can effectively capture both low-frequency and high-frequency components of a signal. This makes them well-suited for a wide range of applications, from audio and speech processing to image and video analysis [7].

Recent studies proposed novel multi-scale architectures that incorporate attention mechanisms [1], recurrent connections [10], and unsupervised learning [9]. These approaches have shown promising results in tasks such as speech enhancement, image restoration, and anomaly detection.

Following an extensive literature survey, in this study we propose an initial multi-scale neural model architecture for extracting TNM model parameters ($A$, $\Delta h^*$, $\chi$ and $\beta$) from DSC curves. The multi-scalability is achieved through the opening section of this model. This section works in three parallel independent branches, where each branch implements an extraction module to mine features of different nature from the input, operating at different frequency scales. Additionally, each extraction module splits the six DSC curves to separate branches, and processes each one of the six curves individually. The outputs of all branches are then concatenated, and the resulting signal is processed together by a series of dense layers. The output of the multi-scale neural model consists of four neurons with a linear activation function, where each neuron provides the value of one of the TNM model parameters. The overall architecture, as we implemented it in our experiments, is depicted in Fig. 2. The architecture of the extraction module is shown in Fig. 3. The kernel size of the convolutional layer varies from 4 (the upper module) to 12 (the middle module) to 24 (the lower module). The lower module input is the original set of DCS curves, the middle module input is downsized to one half, and the upper module input is downsized to one third of the original set of DCS curves.

### 2.3 Dataset

In order to get the sufficient data for neural model training, a set of input structure-output structure pairs needs to be prepared. In this case, the input structure is represented by six DCS curves, and the output structure consists of four TNM model parameters ($A$, $\Delta h^*$, $\chi$ and $\beta$). In our study, the dataset is artificially computed using the algorithm for the Tool–Narayanaswamy–Moynihan

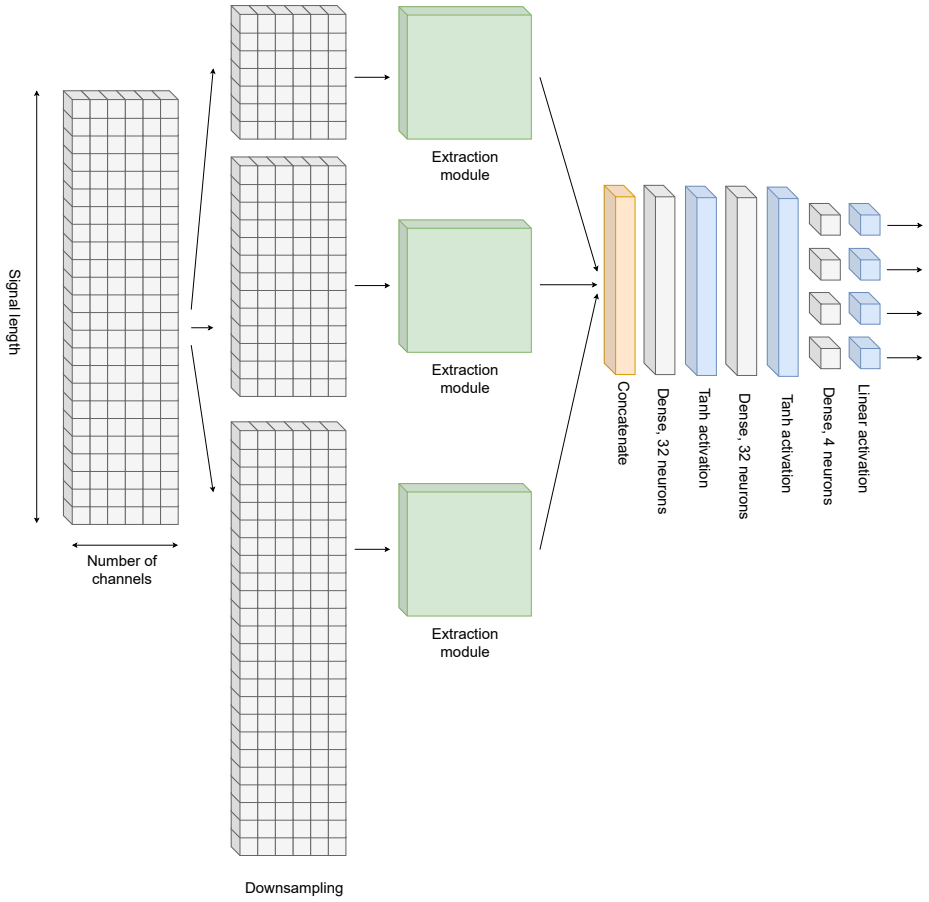


**Fig. 2.** Overall architecture of our implementation of a multi-scale convolutional neural model.

curve-fitting procedure [3]. According to that algorithm, the expression for the non-isothermal steps is represented by

$$T_{f,n} = T_0 + \sum_{j=1}^{n} \Delta T_j \left\{ 1 - \exp\left[ - \left( \sum_{k=j}^{n} \frac{\Delta T_k}{q_k \tau_k} \right)^{\beta} \right] \right\}, \tag{3}$$

where $T_0$ is the initial equilibrium temperature. The fictive temperature $T_f$ in this equation is determined based on the Boltzmann superposition integral over time, which can also be replaced by a corresponding integral over temperature. In practice, the continuous cooling or heating is replaced by a sequence of $n$ temperature jumps $\Delta T$, followed by isothermal holds with duration determined by the cooling and heating rates $\Delta t = \frac{\Delta T}{q}$, as suggested in [3]. The magnitude of

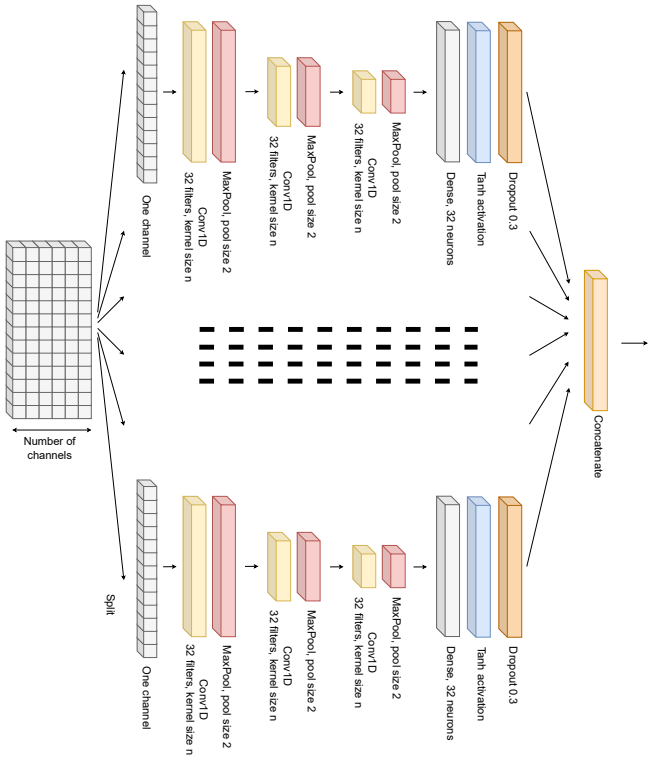


**Fig. 3.** Extraction module. Input multi-channel signal is split into six separate branches, each of them processed individually.

$\Delta T$ must be sufficiently small to ensure linearity. The self-retarding kinetics can be introduced by dividing the aging time into $k$ sub-intervals and calculating $T_f$ and $\tau$ at the end of each. The sub-intervals are usually determined by dividing the aging time into evenly spaced logarithmic intervals. The fictive temperature is then used to calculate the normalized heat capacity $C_{\mathrm{p}}^{\mathrm{N}}$, as follows.

$$C_{\mathrm{p}}^{\mathrm{N}} = \frac{\mathrm{d}T_f}{\mathrm{d}T}. \tag{4}$$

The use of Equations (3) and (4) allows for the implicit determination of DCS curves for a selected course of repeated heating and cooling of the sample. Nevertheless, this process is computationally intensive – calculating for one quartet of TNM model parameters takes approximately 10 minutes on a high-end computer. For the purpose of this study, 5000 sets of DCS curves are computed. Each parameter of the TNM model for every computed set of DCS curves is selected randomly with a uniform probability distribution. The ranges of each parameter are defined on the basis of naturally occurring values in real amorphous materials, and are listed below.

– $\log_e A$: [-18; -630]
– $\Delta h^*$: $[2 \times 10^5; 12 \times 10^5]$ $\mathrm{Jmol}^{-1}$
– $\chi$: [0.2; 1]
– $\beta$: [0.2; 1]

Therefore, 5000 elements of input structure-output structure pairs are prepared to train the multi-scale convolutional neural model. Each input structure contains six-channel signal of length 18501. Each output structure is compounded of four values: $A$, $\Delta h^*$, $\chi$ and $\beta$. These elements are randomly divided into training and test sets in a 4 : 1 ratio. Six selected representatives are depicted in Fig. 4. Note that, for the purpose of training the neural model, the values in the output structure are transformed to the range of $[-1, 1]$. This is done to ensure that all values are given equal weight in the cost function.

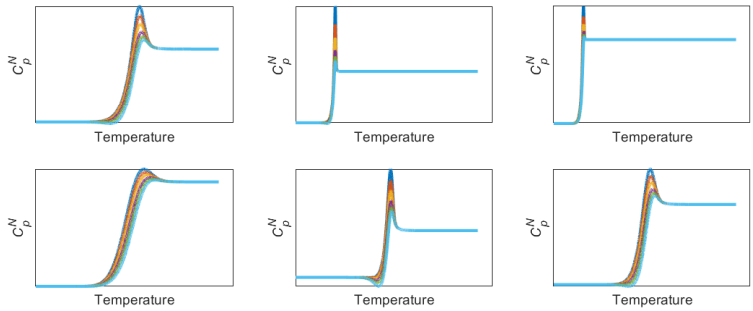


**Fig. 4.** Examples of DCS curves used in the dataset depending on the selected TNM model parameters.

## 3 Experiments and results

### 3.1 Training details

The SGD algorithm is selected as an optimizer, since it is generally considered to provide acceptable performance in most of the cases - it is efficient, it provides a good generalization capability, it is scalable to large datasets, and last but not least, it is simple to implement [2]. Mean Square Error loss function is used to evaluate training process and Mean Absolute Error function is applied to validate the results. Each training experiment was performed five times due to a stochastic character of training. This is not a significant enough number of repetitions for statistical analysis; however, the time constraints of the experiments did not allow for more repetitions. The instance, which performs best over training set, is selected for further validation. All the parameters of the training are summarized in Table 1.

**Table 1.** Parameters of training

| | |
|---|---|
| Input shape | 18501 x 6 |
| Training algorithm | SGD algorithm |
| Loss function | Mean Square Error |
| Validation metric | Mean Absolute Error |
| Number of training experiments | 5 |
| Maximum epochs | 500 |
| Stopping criterion | Maximum epochs reached |
| Learning rate $\alpha$ | 0.01 |
| Batch size | 2 |

### 3.2 Evaluation of multi-scale neural model

The trained neural model is evaluated in terms of Mean Square Error and Mean Absolute Error. The resulting values are summarized in Table 2. Note that the values transformed to the range of $[-1, 1]$ are used to determine the metrics, in order to keep the equal weight of each value to the metric.

**Table 2.** Results of neural model evaluation

| Data | Mean Square Error | Mean Absolute Error |
|---|---|---|
| Training set | 0.000337 | 0.0131 |
| Test set | 0.00189 | 0.0252 |

The resulting Mean Absolute Error of the model over the test set is found to be 0.0252, indicating a high level of accuracy in the predicted values. Since the transformed output values are observed to fall within the range of $-1$ to 1, the resulting model output error is below two percent. These findings suggest that the proposed neural model may be effective in accurately extracting the desired values from the input data. In order to demonstrate the functionality of the model, Fig. 5 shows a plot of the randomly selected input set of DCS curves and the associated set of curves calculated using the output parameters of the model presented in this study. These parameters are summarized in Table 3.

**Table 3.** Expected and predicted outputs from neural model used for comparison in Fig. 5

| Parameter | Expected value | Predicted value |
|---|---|---|
| $\log_e A$ | -120.10 | -122.92 |
| $\Delta h^*$ | $1.0300 \times 10^6$ | $1.0294 \times 10^6$ |
| $\chi$ | 0.3600 | 0.3484 |
| $\beta$ | 0.3400 | 0.3378 |

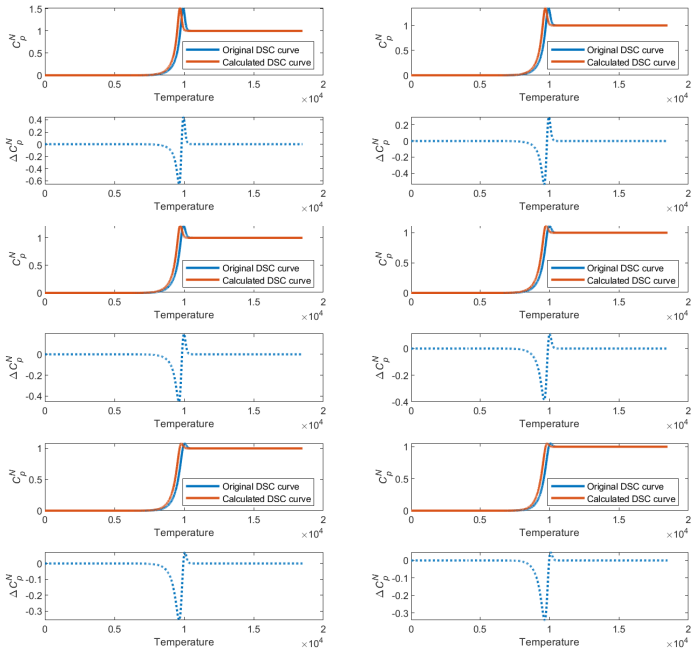


**Fig. 5.** Example of the original set of DCS curves and DCS curves computed by four parameters provided by neural model presented in this study.

Moreover, it is possible to notice a rather large difference in the accuracy of the model on the training set and on the test set. Although this phenomenon often indicates overfitting of the model, in this case the cause is more likely to be due to the insufficient range of the training set. 5000 elements for a four-dimensional space probably does not offer enough data power to fully map the solved problem.

## 4 Conclusion

Glass transitions are a crucial phenomenon in the field of amorphous materials with potential for use in various applications. The TNM model is widely used to describe the enthalpy relaxation behavior of amorphous materials. However, determining the appropriate values for its parameters can be challenging due to their interdependence and variation based on material and experimental conditions. We propose a multi-scale convolutional neural model that can accurately predict the TNM parameters of glass-forming materials directly from the set of their differential scanning calorimetry curves. Our results show that

the proposed neural model can effectively extract the desired values from the input data with high accuracy. However, the insufficient range of the training set may have caused the discrepancy between the accuracy of the model on the training set and the test set. Hence, in future work we will focus on working with a significantly larger dataset and also focus on optimizing the architecture of the proposed neural model.

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