Hybrid Mechanistic Neural Network Modelling of the Degree of Cure of Polymer Composite

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Abstract: A hybrid mechanistic/neural network model was developed for the industrial polymer composite curing process of a fibre-reinforced polymer composite. A hybrid model with parallel scheme and a hybrid model with the combination of series and parallel schemes were developed. It is found that the hybrid model with the combination of series and parallel schemes gives better performance. It is shown that the developed hybrid model is more accurate than its mechanistic and neural network counterparts in predicting the degree of cure based upon the temperature and time data. The hybrid model is 7.7% and 17.1% more accurate than the neural network model and the mechanistic model respectively in terms of sum of absolute errors.

1 INTRODUCTION

Fibre-reinforced polymer composites (FRPCs), or simply composites, are materials that consist of two phases: the matrix phase, delivered by a tough but structurally weak thermoset resin, and the reinforcing phase, delivered by filaments (diameter ~10μm) of a strong and stiff material (Smallman & Bishop, 1999). This combines the properties of both constituents (Taj et al., 2007) and gives the final composite material with high stiffness to weight ratio and improved strength in comparison to other structural materials (Ahmad et al., 2021). Common fibres used in modern composites include carbon, boron, glass, aramid and naturally occurring plant fibres. The fibres are impregnated with the matrix phase, setting the fibres into place and providing lateral support, while also minimising damage to the composite by providing plastic deformation characteristics lacking in the reinforcing phase (Soutis, 2005).

Originally developed for the aerospace industry to reduce the weight of aircraft (Ahmad et al., 2021), but due to the reduction in price of composites in recent years, many other applications have been found. Aerospace now only accounts for 20% of the carbon fibre market (Soutis, 2005). The same properties that make composites appropriate for use in aeroplanes make them useful for increasing fuel efficiency in modern cars. Roughly 75% of fuel consumption of a car is directly related to its mass and, thus, lightweight composite panels have been the material of choice for hybrid and battery powered cars to maximise their driving range, and on high performance supercars to increase performance. Due to the energy absorption of an epoxy matrix phase, composite materials provide increased passenger safety when compared to metal components, and now composites are used to create entire cabs for heavy trucks and large panels on buses (Friedrich & Almajid, 2013). This study uses data from an automotive industry composite curing process.

Composites are also used extensively outside of transport applications. Composites have almost fully replaced conventional materials, particularly in conjunction with ceramics (Grand View Research, 2019), in the ballistic armour industry. Polymer composite materials are also finding extensive use in civil construction, particularly in repair and rehabilitation of existing concrete structures (Pendhari, et al., 2008) (due to strength and toughness). Fibreglass is an incredibly common material in the building of yachts and high performance dinghies (to reduce weight and increase speed). Polymer and ceramic composites are being introduced into systems involving corrosive chemical storage (due to the chemical resistive nature of the polymer matrix phase).

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In the manufacturing of epoxy composites and other thermosetting resins, a technique called resin transfer moulding (RTM) is used (Liu et al., 2019). This is where the matrix-phase monomer resin is applied to the reinforcing phase already in the mould, mixed with a curative, usually an amine or anhydride (Hara, 1990), to harden the resin by polymerisation around the fibre structure in the shape of the mould. The temperature control of the curing process is delivered by placing the mould in an oven and the energy supplied to the part activates the exothermic curing reaction. The temperature profile of the reaction is dependent on the heat produced and the energy supplied to the reaction by the mould system, which can be used to control the degree of cure (Joshi et al., 1999).

However, improper curing via incorrect temperature control can cause irregularities in the final mechanical properties of the composite. A key parameter in the strength of the composite is the strength of the bond between the fibre and the polymer, and due to the difference in coefficients of thermal expansion between the two phases, the incorrect heating of the composite during cure can lead to residual micro-stresses in the composite structure after the cure has been completed (Kondyurin, et al., 2012). Thus in order to exert optimal control over the final properties of the composite, accurate models of the curing process must be created.

Models for degree of cure in polymer composite moulding processes can be generally classified into two catagories: mechanistic models and data-driven models. Mechanistic models are based on first principles such as reaction kinetics. They should be accurate and reliable if precise mechanistic knowledge is available. However, some mechanistic knowledge can be complex and only partially known. In such cases simplifications and assumptions have to be made leading to reduced model accuracy. Furthermore, the development of mechanistic models are typically time consuming and effort demanding. Data-driven models can be developed quickly and can give accurate predictions when used within the range covered by the training data. However, they are of black box nature and are difficult to interpret. They can also give large errors when applied outside the range covered by the training data. A hybrid model combining both mechanistic model and data-driven model could exhibit the advantages of both types of models.

The most common technique to model the cure of polymer composite is the use of semi-empirical mechanistic modelling. These models state a general order for the reaction process replacing the concentration of the present species in the kinetic equation with a measure of the degree of cure (Halley & Mackay, 1996). The model parameters are found via experimentation much like that of first-principle mechanistic modelling. Simple semi-empirical models were used by (Karkanas et al., 1996) and (Du et al., 2004) for modelling a composite curing process, and both managed to produce models that were accurate for ~80% of the experimental data. However, these models did not have consistent reaction orders as the temperature changed and required other equations, such as the diffusion factor used by (Du et al., 2004) to manipulate the reaction rate constant in the latter stages of the reaction. To improve the areas of poor accuracy, first principle models can be used, such as those developed by (Blanco et al., 2005) and (Riccardi et al., 2001), which provide a consistent reaction order for the system that does not change with the temperature, but the accuracy is still only acceptable for ~80% of the cure process, thus not justifying the added complexity of these models. Alternatively, Joshi et al. (1999) used two separate semi-empirical models to model their composite curing process, with the Arrhenius parameters and reaction order changing after degree of cure reaching 0.18, but there were significant inaccuracies in this investigation at the boundary between models despite the model being accurate at the beginning and end of the cure process.

Data-driven models, in particular neural network (NN) models, have been reported for the modelling of degree of cure in reactive polymer composite moulding processes. Zhang & Pantelelis (2011) developed a bootstrap aggregated neural network model that predicted the electrical resistance of a polymer/carbon composite part during curing and used this to predict the degree of cure. The one-step ahead model used for effective process optimisation which increased the maximum degree of cure for a part by as much as 0.2 in offline optimisation. Similar results were found from the model produced by Lee & Price (1996), who modelled the curing of epoxy by a NN that directly predicted the degree of cure rather than resistance. It was found that the NN model was more accurate when predicting degree of cure (DOC), with the absolute error consistently lower (< 0.04) than that of the analytical model (≤ 0.12). What is observed is, similar to that found by Zhang & Pantelelis (2011) that the NN model tended to underpredict the degree of cure as the curing neared completion ($\alpha > 0.8$) opposed to the analytical model which overpredicts. In addition to this, Su et al. (1998) found that their NN models for controlling a curing process exhibited poor adaptability when the system to be modelled lies outside of the training data limits, a problem that was not apparent with analytical models.

Hybrid mechanistic/neural-network models (HNN) use a mechanistic component and a NN component to strike the balance between the speed and accuracy of a neural network and the applicability of a mechanistic model (i.e. allowing the HNN to operate outside of the range of training data effectively). To the authors' knowledge, there are no known examples of applying a HNN to the curing process of reactive polymer composite moulding, but in other complex systems, HNNs have found successes. Lee et al. (2002) developed 4 models for analysis of the treatment of coke-plant wastewater (mechanistic, NN, serial-HNN, parallel-HNN). The serial-HNN used a single NN to estimate the parameters for the two types of biomass which fed into the mechanistic model, and the parallel-HNN model used a single NN to produce error estimations for the mechanistic model to be combined to guide the mechanistic model output. The parallel-HNN was the most accurate of the two HNNs investigated, and while the NN outperformed the parallel model in training but for the validation on unseen data, the hybrid model was more accurate. This shows the advantage of using a hybrid model, in that it has greater ability to estimate unseen data than a standard neural network or mechanistic model. Tian et al. (2001) applied a parallel hybrid neural network model to a polymerisation process of methyl methacrylate. In the reported study, rather than one neural network, a stacked neural network (bootstrap aggregated neural network) was used to predict the error from the mechanistic model to compare to using a single optimised neural network for the parallel hybrid. The stacked neural network was more successful, but most interestingly the confidence bounds (used as an indication of reliability on unseen data) were incredibly tight for the parallel model's prediction of conversion.

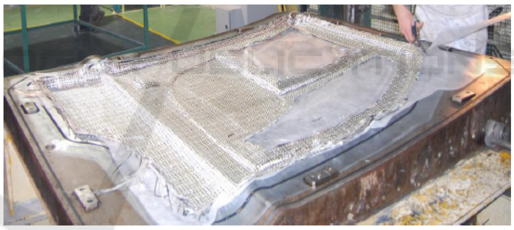
This paper presents a hybrid mechanistic and neural network model for the modelling of degree of cure in an industrial polymer composite moulding process.

2 MODELLING OF DEGREE OF CURE

2.1 Data Collection

The cure-process raw data was provided by SOTIRA, a subsidiary of the SORA Composites Group, who manufacture plastic/carbon composite parts for the automotive and agriculture industries. The raw data consisted of resistance and temperature measurements of the composite part at minute intervals during the industrial manufacturing process, collected using OptiMold from Synthesites (Zhang & Pantelelis, 2011). The temperature was controlled at 114ºC but fluctuated randomly throughout each experiment due to process noise. 25 sets of 19-minute-long runs were provided for this investigation (labelled A1-A25). Figure 1 shows the pictures of the product and the mould.

(a)



(b)

Figure 1: Product (a) and mould (b) for the reactive polymer composite moulding process.

The degree of cure (DOC) was determined using Eq(1) (Zhang & Pantelelis, 2011). The value of α_{max} was calculated for each data set using Eq(2), where $R_{max,O}$ represents the maximum resistance achieved by any of the 25 experiments (4.05×10^6 MΩ on experiment A6). This is considered universal (A1-A25) as each composite part is assumed to be the same shape and size. However, R_{min} was taken on a case-by-case basis as due to the confidential nature of the experimental setup, it is assumed that the resin and the curative have not interacted before the experiment begins.

$$\alpha_E = \frac{\log(R_i) - \log(R_{min,i})}{\log(R_{\max,i}) - \log(R_{min,i})}\alpha_{\max,i}\,, \quad i = \{1, \dots, 25\} \tag{1}$$

$$\alpha_{\max,i} = \frac{\log(R_{\max,i})}{\log(R_{\max,O})}\,, i = \{1, \dots, 25\} \tag{2}$$

where α_E is the experimental DOC, α_{max} is the maximum DOC, R is resistance (MΩ), and i is experiment number.

Each model development process consisted of fitting and testing. The experimental data was divided into two groups: 70% for fitting and 30% for testing. This was the ratio used in (Zhang & Pantelelis, 2011) when building their NN from a similarly sized dataset using degree of cure data, resulting in an accurate model. The fitting data was used to define the model parameters (i.e. kinetics constants or node weights) where the experimental DOC data was available to the model. The testing data set was used to test the model to get an indication of how the model will perform when it is applied to the real life cure process, i.e. testing performance as opposed to "recall" (Lee et al., 2002). 7 runs (~30%) were selected at random for use in the testing stage. These were: A2, A3, A12, A13, A14, A21 and A23, and Figure 2 shows how the spread of fitting and testing data compares.

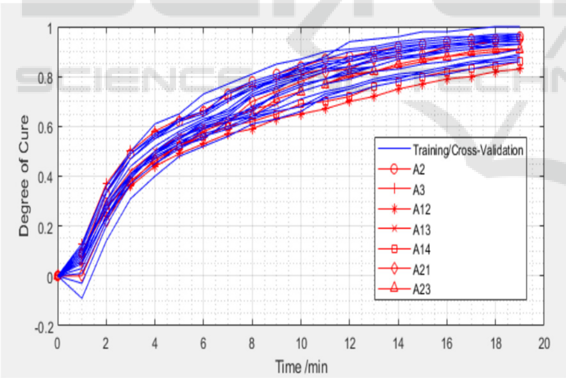


Figure 2: Degree of cure in training and testing data.

For the NN and HNN models, the fitting group was further divided again into training and cross validation groups. The training group is used to define several combinations of node weights that model the input/output relationship adequately. These candidates are then exposed to the cross-validation set which allows the most accurate model to be chosen from the candidates. This is not to be confused with testing, as the output data is still available to the model, and the candidate selected will carry any biases present in the cross-validation data (Demuth & Beale, 2004).

2.2 Mechanistic Modelling

A mechanistic model was identified to act as the mechanistic component of the hybrid neural network (HNN) model, but also to be used for comparison purposes to the HNN model to test model performance. To identify the best mechanistic model, three types of semi-empirical models were identified and tested to compare their respective accuracy and precision. The three mechanistic models are shown in Eq(3) to Eq(5) and are labelled as Models 1 to 3. These are the semi-empirical models presented by (Karkanas et al., 1996) that apply to the majority of different matrix-phase curing processes, and versions of these models are used by Joshi et al. (1999) and Du et al. (2004).

Model 1:

$$\frac{\mathrm{d}\alpha}{\mathrm{d}t} = k_1(1-\alpha)^n\,, k_1 = A_{0,1}e^{\frac{-E_{A,1}}{RT}} \tag{3}$$

Model 2:

$$\frac{\mathrm{d}\alpha}{\mathrm{d}t} = (k_1 + k_2\alpha^m)(1-\alpha)^n\,, \quad k_x = A_{0,x}e^{\frac{-E_{A,x}}{RT}} \tag{4}$$

Model 3:

$$\frac{\mathrm{d}\alpha}{\mathrm{d}t} = k_1\alpha^m(1-\alpha)^n\,, k_1 = A_{0,1}e^{\frac{-E_{A,1}}{RT}} \tag{5}$$

In the above equations, k_1 and k_2 are reaction rate constants (min^{-1}), m and n are reaction orders, A_0 is the nominal Arrhenius pre-exponential factor (min^{-1}), E_A is the nominal activation energy (J.mol^{-1}), and R is the universal gas constant (J.mol^{-1}.K^{-1}).

The model parameters in these equations are obtained by fitting the training dataset. This was carried out by using the FMINCON function from the MATLAB Optimisation Toolbox. This is a non-linear optimisation algorithm that locates points of zero gradient in the objective function based on several inputs. For larger optimisation problems, a Hessian is used, and for small/medium size problems a Sequential Quadratic Programming method is used to find the optimum (Novac et al., 2009). While FMINCON finds only local minima (therefore the output objective function is dependent on the initial guess values) and genetic algorithms have been found to be more accurate optimisers. The fitted mechanistic models were then applied to the testing dataset with the results given in Table 1. It can be seen that Model 3 was identified to be the most accurate mechanistic model having the lowest SAE (sum of absolute errors) and SSE (sum of squared errors). Model 3 is used in the development of HNN model.

Table 1: Identified model parameters and performance on the testing data.

	Model 1	Model 2	Model 3
$A_{0,1}$	9.997×10^4	9.928×10^4	9.996×10^4
$A_{0,2}$	-	9.997×10^4	-
$E_{A,1}$	4.297×10^4	5.334×10^4	4.900×10^4
$E_{A,2}$	-	4.164×10^4	-
m	-	0.2119	0.2033
n	1.0893	1.3787	1.3728
SAE	6.695	6.658	6.636
SSE	0.527	0.521	0.520

2.3 Neural Network Modelling

A neural network model was developed for the purpose of comparison with the hybrid model. The model inputs are curing temperature (T) and curing time (t). A two hidden layer feedforward neural network is developed. The fitting data were further portioned to training data (75%) and validation data (25%). The number of hidden neurons were determined by considering a range of hidden neurons and the one giving the best performance on the validation data is considered to have the appropriate network structure. For building the NN model and the NN part of the HNN model, the MATLAB Deep Learning Toolbox was employed. This allows the user to specify the architecture of different inbuilt neural network systems. The neural network that was used for this investigation was the feedforward neural network as shown in Figure 3. The feedforward neural network architecture is simple and does not require time delays or for the neural network to be recurrent which allows greater control for the user and for a greater number of network types to be investigated. The final selected NN model is shown in Figure 3, where the numbers of hidden neurons were determined through cross-validation using the validation data.

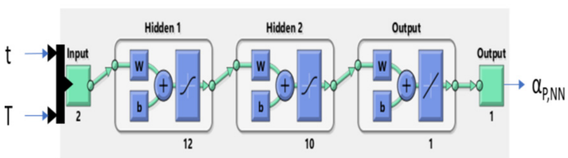


Figure 3: NN model structure.

3 HYBRID MODEL

3.1 Model Structure

The hybrid neural network is shown in Figure 4. The neural network model was trained to model the error, α_{Error}, between the mechanistic model and the experimental value for α calculated using Eq(1) and Eq(2). This does not exactly follow the explicit parallel form of the hybrid created by (Lee et al., 2022) as their hybrid relied on inputs for the mechanistic model and the neural network being the same with no series characteristics. The model setup following this mantra is shown in Figure 4, however initial modelling studies found greater accuracy using the output of the mechanistic model as the input for the neural network. Hence, the model shown in Figure 5 was preferred over Figure 4.

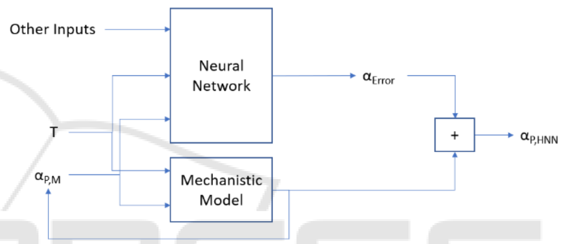


Figure 4: Hybrid model with the parallel scheme.

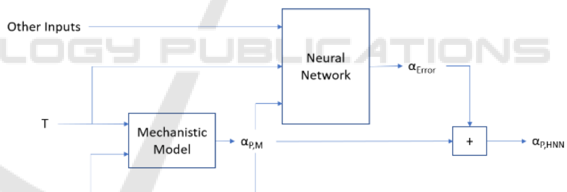


Figure 5: Hybrid model with the combination of series and parallel schemes.

3.2 Model Performance

Table 2 shows the performance of the mechanistic model, NN model, and HNN model on the testing data. It can be seen from Table 2 that the mechanistic model gives the worst performance. This could be due to that the three considered mechanistic models do not fully represent the reaction kinetics of the reactive polymer curing process. The NN model gives better performance than the mechanistic model in this case. This could be due to the excellent capability of NN in representing nonlinear functions. The HNN model integrating a mechanistic model and an NN model gives the best performance. In terms of SAE, the HNN model is 7.7% more accurate than the NN model and 17.1% more accurate than the mechanistic model. Figures 6 to 12 show the predications of the three models on the testing batches.

Table 2: Model performance on the testing data.

Models	SSE	SAE
Mechanistic	0.5197	6.6356
NN	0.3806	5.9606
HNN	0.3741	5.5011

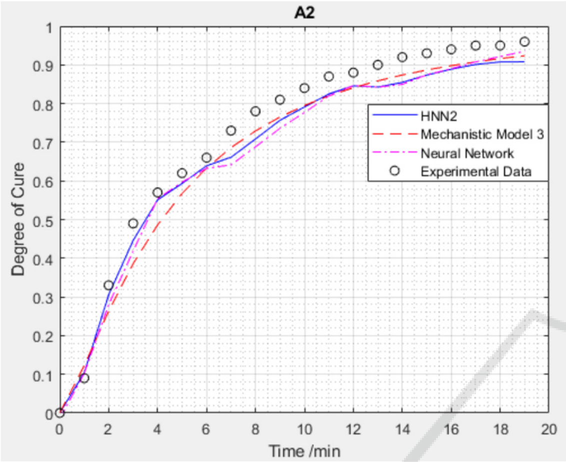


Figure 6: Model predictions on the test batch A2.

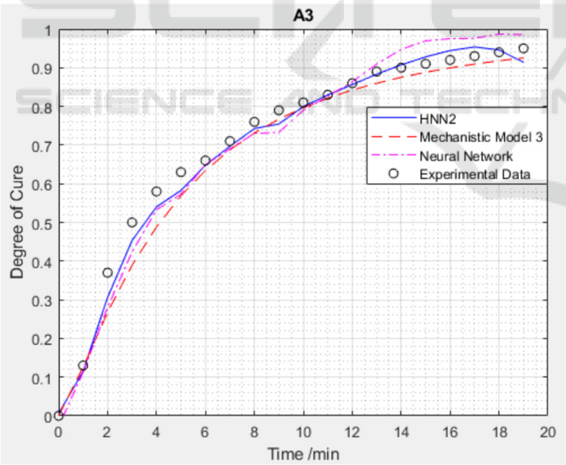


Figure 7: Model predictions on the test batch A3.

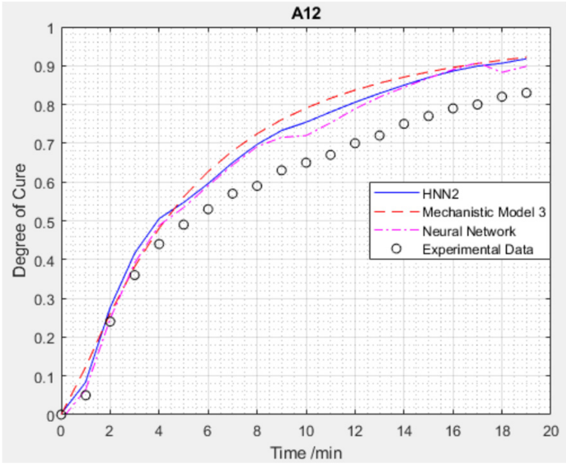


Figure 8: Model predictions on the test batch A12.

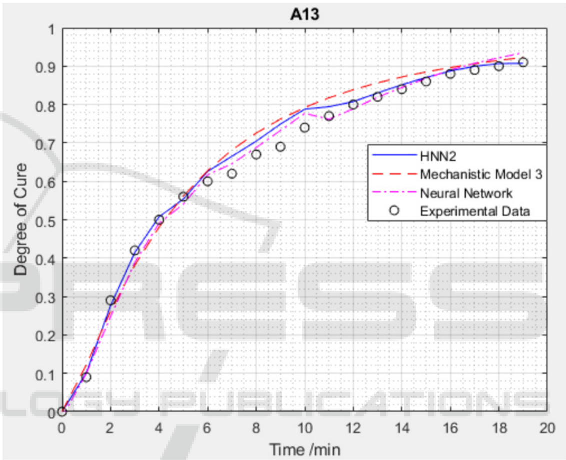


Figure 9: Model predictions on the test batch A13.

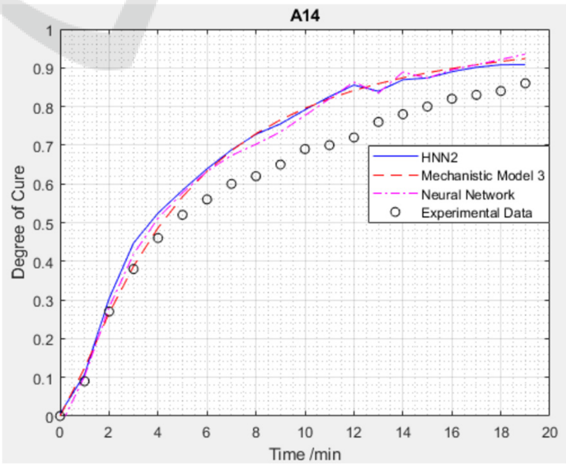


Figure 10: Model predictions on the test batch A14.

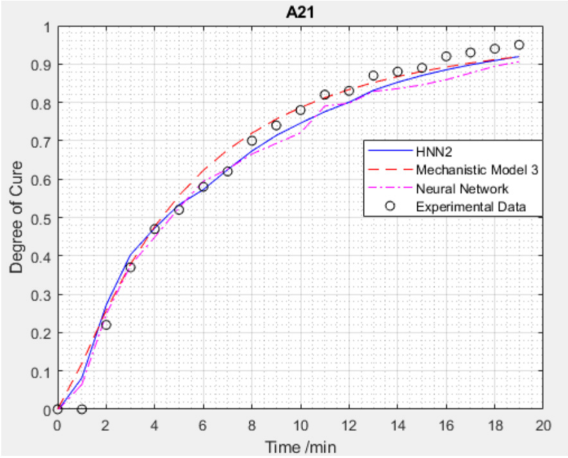


Figure 11: Model predictions on the test batch A21.

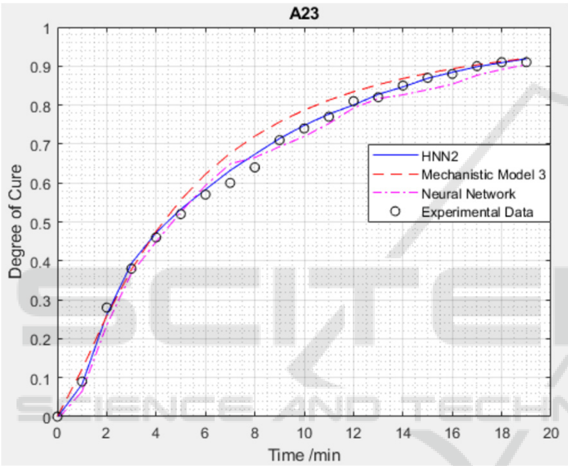


Figure 12: Model predictions on the test batch A23.

4 CONCLUSIONS

Hybrid mechanistic/neural network models for the curing of FRPCs are developed in this paper. The obtained results have shown that the hybrid model with the combination of series and parallel schemes gives the best performance and it can provide 7.7% better accuracy than the NN model and 17.1% more accurate than the mechanistic model in terms of sum of absolute errors, using only curing temperature and curing time as model inputs. An important factor for increasing the accuracy of the HNN was found to be high data-diversity in the cross-validation training group, as well as mechanistic-component accuracy. When the mechanistic model is improved, and the fitting data set is large and diverse enough, the HNN can achieve further improved prediction performance.

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