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Research Article

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Posted Date: October 1st, 2021

DOI: <https://doi.org/10.21203/rs.3.rs-952869/v1>

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Neural Network Modeling of NiTiHf Shape Memory Alloy Transformation Temperatures

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Abstract

Machine Learning (ML) techniques enabled the effective correlation of alloy characteristics. In this paper, an ML Neural Network (NN) model is proposed to predict the transformation temperatures (TTs) of NiTiHf for a given combination of elemental composition and processing parameters within an acceptable confidence interval. The main challenge in NiTiHf TTs prediction is the high dimensional dependency of TTs to various parameters as well as not fully clear governing physics. Considerable experimental research was performed from 1995 to design the NiTiHf TTs which in turn paved the path to feed the ML algorithms which are now presenting an excellent approach for material design. A thorough data set is constructed from both unpublished data and available literature and then analyzed to select twenty input parameters to feed the NN model. To model and forecast the TTs of NiTiHf with a wide range of TTs up to 800°C, a total of 173 data points are gathered, verified, and selected. The model's overall determination factor (R^2) was 0.92, indicating the viability of the proposed NN model in showing the link between material composition, processing factors, and then defining the TTs of NiTiHf alloy. The effort additionally validates the generated results against existing data in the literature. The validation confirms the significance of the proposed model.

Keywords: NiTiHf Shape Memory Alloys, Neural Network, Machine Learning, Data-driven Material Design, Computational Material Manufacturing

List of acronyms

Af	Austenite Finish Temperature
Ap	Austenite Peak
As	Austenite Start Temperature
HTSMAs	High-Temperature Shape Memory Alloys
LPBF	Laser Powder Bed Fusion
LTSMA s	Low-Transformation Temperature Shape Memory Alloys
Mf	Martensite Finish Temperature
ML	Machine Learning
MLP	Multilayer Perceptron
Mp	Martensite Peak
Ms	Martensite Start Temperature
NN	Neural Network
SMA s	Shape Memory Alloys
SDA	Suspended Droplet Alloying
TTs	Transformation Temperatures
TH	Transformation Hysteresis

1. Introduction

1.1. Machine Learning approaches for alloy design

There is a current challenge to develop an inclusive approach to model and formulate the material design and behavior. ML approaches have the potential to significantly decrease the need for complex physical and mathematical considerations(Ref 1), as well as, the number of physical experiments needed to optimize the process of materials design and discovery(Ref 2,3). In addition, the ability for efficient, accurate predictions and performing complex pattern recognition and regression analyses, made ML approaches good candidates in the area of material design and discovery(Ref 4–8).

ML is currently implemented to design new single-phase materials in many areas like thermoelectrics(Ref 9), semiconductors(Ref 10), superconductors(Ref 11), metallic glasses(Ref 12), inorganic crystals(Ref 13), superconductors(Ref 11), Heusler compounds(Ref 14) and more. However, these designs focus on single-phase materials and disregard the effect of process variations and microstructure on the material's behavior. The majority of alloys are consisting of several elements with inclusions and impurities that are originated from the manufacturing or post-processing steps that alter their performances(Ref 15). The recent study of material compositions and post-processing effects is immature. Heat treatments and mechanical post-processing are an essential part of material development to create a multi-phase structure reaching desirable performances. This suggests an enormous open space for material design through ML approaches to search and build an alloy design. The data source to feed the ML models are the data gathered from the literature (Ref 6,16–21), data from the theoretical models (Ref 22), data simulated from computational (Ref 23), and the data created by physical experiments or a combination of these sources. The most trustable data to feed the ML algorithms are the experimental ones as they are indeed tested and measured. The present work is focused on gathering the experimental data from literature and also through physical experiments. Fig. 1 represents the general approach for data-driven ML material design.

Among ML algorithms, Neural Network(NN) is the most widely used model due to its ability to handle large datasets that can be available in addition to its strong computational power and sophisticated algorithm architecture (Ref 8). NN methods proved their validity in material design and optimization areas(Ref 24,25,34,35,26–33), and therefore, the present work is focused on implementing the NN method, as well.

1.2. SMAs and TTs design parameters

Shape memory effect and superelasticity are the two principal features of SMAs. These effects are resulting from a reversible microstructure transformation and rely on the phase transformation between austenite and martensite phases that occur through applying stress or changing the temperature(Ref 36,37). These effects attracted many industries to develop a wide range of applications from biomedical to aerospace applications(Ref 38–41).

The phase TTs consist of four critical temperatures; Martensite Start (Ms), Martensite Finish (Mf), Austenite Start (As), and Austenite Finish (Af) temperatures. Ms presents the temperature at which the transformation from austenite to martensite starts, while Mf represents the temperature for complete martensitic transformation. The As and Af indicate the phase change from martensite to austenite as a result of heating (Ref 42). Martensitic Peak (Mp) and Austenitic Peak (Ap) represent temperatures with maximum cooling or heating rate during the material thermal cycling, respectively. Transformation hysteresis (TH) is an important design factor that is defined as the difference between Af and Ms or Mp and Ap that presents the SMA material thermodynamic efficiency. TH represents the amount of energy that material dissipates during each cycle.

NiTi, as the most used SMAs, is showing shape memory effect and superelasticity that makes it a suitable candidate for many applications. However, due to the limited range of the transformation temperatures that cannot exceed 120 °C, the binary NiTi alloys are not suitable for applications having high working temperatures (Ref 39,43–47). Many studies in the literature investigated adding various elements such as Zr (Ref 48), Au (Ref 49), Pd (Ref 50), Pt (Ref 51), Ce (Ref 52), Er (Ref 53), and Hf (Ref 54) mostly to increase the TTs (Ref 55) or Co, Cr, V, Fe and Mn to reduce the TTs(Ref 56). Considering the challenges of adding the third alloy, Au, Pd, and Pt are very expensive alloys and Zr increases the risk of oxidation in the material (Ref 55,57). There are also some limited studies on the addition of Ce or Er (Ref 53).

Adding Hf as the ternary element to NiTi alloy has proved to be promising as the next generation of SMAs. This alloy shows good mechanical properties at a relatively low cost, offers a very wide range of TTs, and offers the possibility of higher strengthens for repeatable functional performances via heat treatments and without any cold work. NiTiHf alloys have been studied mainly as high-temperature SMAs (HTSMAs) (Ref 54); however, based on the elemental composition, they can offer low transformation temperature SMAs (LTSMAs), as well (Ref 58).

The elemental composition of NiTiHf alloys is the main factor affecting the TTs (Ref 59). It has been shown that for Hf content of less than 10%, depending on the NiTi content, the alloy can show medium or very low TTs ranging from about -140 °C to 100 °C (Ref 58). Increasing the Hf content by more than 10% leads to an increase in the TTs up to 644 °C (Ref 54,60). The TTs of NiTiHf alloys are also affected by the Ni content; Ni-rich alloys (i.e. Ni% more than 50%) have lower TTs (Ref 54,61).

Heat treatments such as homogenizing, solutionizing, annealing, and aging, and mechanical post-processing like hot rolling, cold rolling, and precipitation hardening are the most common methods implemented to regulate the properties, including the TTs, of the SMA alloys, besides the chemical composition (Ref 59,62–67). These processing approaches are mostly employed to improve the

thermo-mechanical properties of the NiTiHf alloys(Ref 59), with a significant effect on TTs (Ref 39,68). Various studies experimentally investigated the effect of heat treatments or mechanical works on the NiTiHf. While aging is the most experimentally studied heat treatment to control the TT variation of NiTiHf (Ref 69–71), homogenization, extrusion, solutionizing, and additional heat treatments showed to have significant effects on the TTs (Ref 72).

Chemical composition, heat treatments, and metalworking processing are the main parameters affecting the NiTiHf TTs and are the focus of the present work.

1.3. Machine Learning to design the NiTiHf TTs

Almost all the studies have used the experimentation methods to some extent to investigate the effect of various compositions, heat treatments, mechanical treatments, and other factors on the properties of the NiTiHf TTs (see Appendix-A) or its thermomechanical behavior (Ref 73,74). Experimental methods are costly and time-consuming and may not fully cover the search space when it comes to alloy design.

Correlating NiTiHf TTs with various post-processing parameters and elemental composition stresses the need for comprehensive modeling techniques. Such techniques should be able to address the dynamic nature and multilevel relations between the material inputs, fabrication methods, heat treatment procedures, and the final alloy material performance.

While ML approaches are adaptable for various materials categories, they represent an excellent tool for understanding and modeling active materials such as SMA. For active materials, the material modeling includes an extra fold of material behavior due to the unique material response to the heat or external force. Hence, the design space has an extra dimension compared to other material classes. There are previous studies that shed light on the use of various ML systems in designing SMA and the resultant material and response characteristics(Ref 59,75–78) (Ref 79).

A few works focused on using the ML approaches for NiTiHf material to predict the TTs (Ref 59,76,80). NN model is used in predicting the NiTi material properties such as TTs and strain recovery ratio with limited data points (Ref 80), the work focused on relating the four fabrication process parameters, of NiTi material to the strain recovery ratio and TTs. Other ML-developed models (Ref 59), used a physic-informed feature selection approach with a special focus on the material composition characteristics. This work presents a different approach with respect to previous ones as the data gathered is about an order of magnitude larger than (Ref 76,80) and also on the contrary of (Ref 59) the approach of this work is to consider almost all the experimental steps that are historically used in the processing or fabrication the NiTiHf and not necessarily downsizing the involving features even if they present limited effects on TTs. Almost no process parameter is eliminated to predict TTs as in the future works the model is intended to consider other functional parameters of the NiTiHf along the TTs. In fact, the work is aiming to develop a tool to predict the TTs based on how the NiTiHf is actually heat-treated and processed up to now. Besides, a new way of NN feature preparation respect to similar works (Ref 59,76,80) is presented in which 1 or 0 values are applied for certain heat treatments or post-processing that are performed or not performed, respectively.

In the present work, a NN ML model is developed to predict the TTs of the NiTiHf based on given chemical compositions, fabrication processes, heat treatments, and mechanical treatments. As described above, the uniqueness of the presented approach lies in the type of data points, selected features, and developed and employed ML; the data points investigated in this work contains a large amount of information to be related to the six TTs, namely: M_f , M_s , A_s , A_f , M_p , and A_p . The work targeted only experimental data as the most reliable type of data to feed ML algorithms.

No data from any kind of modeling approach are collected to avoid the presence of any erroneous input. The data is gathered almost from all the laboratories working on NiTiHf all over the world (see Appendix-A). The results and discussion part of related studies are carefully evaluated to perform the feature selection and to obtain a total of 173 data points. Finally, the NN model was further validated for several cases by comparing the predicted values with the experimental works.

2. Methodology

A NN model is developed in the present study to predict NiTiHf TTs. The data set preparation, features selected, ML algorithms, and the error metrics are presented next.

2.1. Datasets gathering and extraction

The development of ML models requires gathering, aggregating, and validating data points that are extracted from the experimental studies on NiTiHf, the used experimental data points used in this work are presented in Appendix-A. More experimental works are studied and a preliminary data set with more than 250 data points are gathered. The data points were then reduced further by feature selection to obtain a total of 173 data points for this study. The feature selection is discussed in the next session.

2.2. Material-process parameters identification and selection

Table 1 represents the features, their related values, and descriptions. A feature selection is implemented by conducting sensitivity analysis combined with expert knowledge on the gathered experimental data points to decide the most important features on NiTiHf TTs variation. To enable the NN model to represent the features, the data set is validated following a set of rules:

- In the data set where M_p or A_p are not reported, the mean value of the M_s and M_f or A_s and A_f is considered, respectively.
- The data set is eliminated if it has more than two missing values.
- The effects of size, stress level, re-melting, oxygen level, mechanical or thermal fatigue over the material are usually not reported in the experimental studies and therefore are neglected.
- Conventional manufacturing methods, arc melting (Ref 81), induction melting (Ref 69), and plasma torch melting [24], are mainly considered in this study. Materials experiencing the same time-temperature profile result in similar TTs. This allowed for the elimination of these conventional approaches from the features. Suspended Droplet Alloying (SDA) as a novel manufacturing method has also been considered in (Ref 82,83). This method showed to increase the TTs and considered as one input. Melt spinning is another manufacturing method assumed to be a combination of conventional melting and extrusion (Ref 84).
- The re-melting process is performed almost always in the fabrication of NiTiHf material. Some studies have repeated the re-melting process several times (Ref 85) to ensure a good homogeneity. The model does not include re-melting as a feature, while it is a process that must be performed on NiTiHf.

- Any heat treatment ranging between 300-900 °C and with a duration of $1h < t < 3h$ is considered aging. TTs generally increase as the aging time increases (Ref 70,71). However, in some cases, aging can lead to the TTs drop [23]. The cooling step is typically performed by air cooling. Aging temperatures are segmented by increments of 100 °C.
- Hot rolling treatment is generally performed between 700 to 850 °C. Since no significant variations in the TTs have been seen between 700 °C and 850 °C (Ref 69), it is assumed that all hot rolling processes are performed at 800 °C. The model does not consider the percentage of the area expansion and the processing time is assumed to be fixed at 6 minutes. The cold rolling process with various parameters is considered a single feature. The hot upsetting treatment presented in (Ref 85), is assumed to be hot rolling as they almost have the same procedure.
- Annealing temperatures are rounded to the closest hundred (Ref 86).

2.3. Machine Learning Algorithm

2.3.1. Neural network model formation and evaluation metrics

In the present work, a NN was developed by three layers of input, hidden, and output. Each layer has neurons that connect to adjacent layers of neurons by weights and biases. Training the NN was performed by adjusting these values that are updated in each iteration to minimize the loss function between the expected (targets from the data set) and predicted outputs.

A Multilayer Perceptron (MLP) NN network was developed to relate the input and outputs. The sigmoid activation function was implemented for the NN model:

$$\text{Sigmoid}(x) = \frac{1}{1 + e^{-x}} \quad \text{Eq. 1}$$

To evaluate the model, cross-validation (Ref 87,88) and loss functions (Ref 87,88) were employed as the main metrics for model accuracy and model error, respectively. Considering the cross-validation, the data points were randomly divided into three sets; 70% training, 15% cross-validation, and 15% testing. An early stopping approach (Ref 87,88) was considered to stop the model training. The stopping condition was set to update the weights and biases for 50 more iterations after the error continuously increases for the validation data set.

The training was performed based on the overall determination factor, as defined below:

$$R^2 = 1 - \frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{\sum_{i=1}^n (y_i - \bar{y})^2} \quad \text{Eq. 2}$$

Where i is the index of the data point, y_i is the predicted values, $\bar{y}$ is the mean of predicted values and $\hat{y}_i$ is the target values. A model with a high and stable R^2 by varying randomly the data sets is selected as the optimum model. To avoid overfitting, the R^2 values for the training, validation and training sets are set to have a maximum difference of 0.1. The error metric or loss function considered in this work is the Root Mean Square Error (RMSE), as calculated below:

$$RMSE = \sqrt{\frac{\sum_{i=1}^n (y_i - \hat{y}_i)^2}{n}} \qquad Eq. 3$$

The total number of data points is defined by n . RMS determines the amount of dispersion of a set of values from the target value. The μ mean error value is also reported.

2.3.2. NN model setup

A multilayer perceptron NN model was employed using the Levenberg–Marquardt optimization algorithm(Ref 89). Table 1 lists the features and expected NN model output.

- **Data Range and Normalizing:** Considering that the input parameters have different natures and different value ranges, a normalization process was performed to map all the input values in the range between 0 and 1. Values of 1 or 0 are considered if a certain heat treatment or metalworking process was implemented or not implemented on the NiTiHf material, respectively. In NN modeling, it is common to assign the value of zero to the input with no meaningful value (Ref 90).
- **Outputs:** Output parameters are all temperatures. The same nature of the data imposes a low correlation between the features, which is likely preferable to train the NN.
- **Model architecture:** The model training was performed continuously for different architectures, from 1 neuron with one hidden layer to 40 neurons with two layers. For each different model architecture, the R^2 values are measured for all six outputs. A MATLAB code loop ran the model 20 times for each different architecture, meaning different numbers of neurons and different layers to determine the maximum, minimum, and average values of the R^2 for each case. Finally, the optimum architecture was selected.

3. Results

3.1.NN model results

The model architecture is achieved after several iterations to search for the best number of neurons and the hidden layers. The results for the optimum architecture for the NN model are presented in Fig. 2, and the NN model schematic is presented in Fig. 3. The study found that the most suitable number of neurons for the second hidden layer of the NN is 25, as shown in Fig. 2. The neuron numbers in the first layer are 30, and the neuron numbers in the second layer are varying from 23 to 27. Both the minimum and the average of R^2 values in the case of 25 neurons show the highest value for almost all cases and therefore this configuration is set to be a reliable one.

The maximum value of the determining factors, the case of 25 neurons shows high values confirming the result's suitability, as well. That is why a two-layered setting with 30 and 25 neurons in the first and second layer, respectively, is selected as the optimum result. The model was evaluated for other NN settings, with one or two hidden layers; however, all the analyses for the R^2 values resulted in lower values.

Fig. 3 presents the optimized NN model configuration, with two hidden layers and 30 and 25 neurons. The model is updating 930 weights and 61 biases for each iteration to reach the final solution training.

Fig. 4 and Fig. 5 present the training results. The regression plot is presented in Fig. 4 for all the output data points as a united category and each output; As, Mf, Af, Ms, Ap, Mp. The overall R^2 value is 0.92, as shown in Fig. 4a, and every single output is also presenting similar R^2 values ranging from 0.89 to 0.91, factor (Fig. 4b, c, d, e, f, and g). The difference of R^2 values was less

than 0.1 for the training, validation, and testing data sets to guarantee a minimum amount of overfitting. It should be mentioned that the R^2 values related to the training sets are higher than the overall ones. The model shows good results based on the R^2 values, and the data points distribution around the regression line describing the predicted temperature. To better investigate the model's accuracy, the study evaluated a histogram fit in Fig. 5 to assess the abundance of the predicted outputs with certain discrepancies from zero error. The error is defined as the difference between the predicted and target transformation temperatures. The occurrence frequency of the error shows a normal distribution meaning that most of the outputs are well predicted. The mean errors, μ , range between -1.6 to -4.7 °C, and the RMSE ranges from 43.9 to 49.5 °C. Considering that the transformation temperatures ranges of NiTiHf are from -140 °C to 644 °C, the mean error and RMSE are relatively low values and the model shows acceptable results. It is worth noting that TTs discrepancies for SMA material with similar composition and processing in different laboratories showed values higher than the errors of the presented model. For example, Karakoc et al. (Ref 91) and Karaca et al. (Ref 70) processed and extruded Ni50.3Ti29.7Hf20 with homogenization and the TTs showed up to 60 °C discrepancies. The same group further aged the material at 500 °C or 600 °C and the TTs showed up to 40 °C discrepancies. The trained model suggests maximum errors equal or lower than what is reported for similar input in different laboratories and therefore present a successful TTs prediction tool.

The results can be improved by adding more data points to enrich the model leading to higher accuracies. This is possible by a continuous update of the data set by newly released data in the literature.

3.2. Model validation

To verify the performances of the presented NN model, three cases are selected. The data points are selected in a way to have the outputs in at least two references; reference 1 (Ref 91) is out of the data used in the modeling and reference 2 (Ref 70) is involved in the model training. Table 2 presents the selected data points for three cases, the features, and their corresponding TTs based on the references presented. The reason that the TTs for the same selected features are different is due to the variations between different studies.

The model ran 10 times for data points, and all 10 predicted outputs are reported along with the data from the references. The main reason all the 10 prediction results are considered is to highlight the solution area and confirm the robustness of the model against a random selection of the training, validation, and testing data points. Fig. 6 presents the validation results. Considering case 1, the predictions for almost all the iterations have resulted in a value close to the references. The maximum discrepancy is less than 5%. For case 2, also, there is a good agreement between the model and the reference values. In case 2, there are three references considered. In (Ref 70) both the aging treatments with 500 °C and 550 °C are reported and compared with the model results which assumed the aging temperature to be 500 °C for both cases. Considering case 3, the maximum discrepancies show a higher value to the two previous cases, and the discrepancies are less than 15%. The main reason for more discrepancies in case 3, is that the number of data points with similar input variables in the data set is limited leading to a larger error in prediction after the model training. Considering the predicted and the output results, the trained model demonstrates to be an effective tool for the prediction of the NiTiHf TTs based on material processing and fabrication. A mean value of 10 predicted values is shown good agreement with the target data and can be considered a finalized model output.

The model is designed to select the training, validation, and testing data points randomly, and this is the main reason that for each iteration, different predictions are given. The model has been optimized to arrange each training, validation, and test set to obtain the optimal results with the minimum discrepancies. This leads to an accurate prediction, which is very similar to the references. The design and arrangement of the data set for the training, validation, and testing data points are performed to check the possibility of accuracy enhancement. Such predetermined data set arrangements resulted in a possibility of higher accuracies for the present case scenarios. However, NN modeling is dynamic and requires a continuous data set enhancement as the new data points are generated, and the final goal is a robust model being independent of any data arrangement. Therefore, this work does not suggest any predetermined data set arrangement for accuracy enhancement. Alternatively, the effective approach to improve the model performances is to keep adding the newly released data points to the global data set.

4. Conclusion

In this paper, a NN model is developed to predict the TTs of the NiTiHf alloys with different chemical compositions, manufacturing processes, and post-processing treatments. After feature selection, a data set with 173 data points is selected. The model overlooked the effect of some parameters such as oxygen content, various conventional fabrication methods, remelting repetition, size, and the TTs measurement methods as these factors do not significantly affect the predicted TTs compared with the key alloying elements, and also very limited related information is available up to now. RMSE and determination factor evaluations are performed for all and individual output parameters to validate the model. The model is further validated by comparing the predictions with the references outside and inside the data set, which have been used for training and validating the model. Training, validation, and testing sets are arranged in a predetermined way to demonstrate the possibility of more accurate predictions. However, this study does not suggest any data set arrangement to get higher accuracy in NN models. Instead, updating the data points parallelly with the future experimental studies data on NiTiHf alloys is considered one of the most effective approaches to improve the model performances. The developed model showed overall determination factors of about 0.9 for each individual output. The mean errors and the RMSE for each individual output showed to be between 1 to 4.7 °C and 44 to 49.5 °C, respectively. The results show errors equal to or less than the discrepancies of TTs reported with similar chemical and processing parameters in various laboratories. The model, therefore, is well validated. This study offers also the prediction of NiTiHf TH as a major design factor, experienced different processes, or design a NiTiHf alloy system with desired TTs. This work which is presenting a tool able to suggest multi-paths to achieve specific TTs opens the space to tune the input parameters to obtain other desirable material properties. Therefore, such a method can be used to guide the NiTiHf design as it also has considered almost all experimental steps in the processing and/or fabrication that are historically performed on the NiTiHf to tailor the TTs or other functional properties. Future work includes parameter optimization to secure compatible properties with a specific application.

Declarations

Funding

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors.

Conflicts of interest/Competing interests

The authors declare that there are no conflicts of interest that could have appeared to influence the work reported in this paper.

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Tables:

Table 1 Features, range, and description used in the NN model.

No.	Input features	Range	Details
1	Atomic percent of Ni	0 to 100	
2	Atomic percent of Hf	0 to 100	
3	Suspended Droplet Alloying	0 or 1	
4	Homogenization	0 or 1	Temperature: 1050°C, Timing: about 70 h Cooling rate: rapid(Water cooling)
5	Solutionizing	0 or 1	Temperature=1050 °C, Timing: about 1h<t<3h Cooling rate: rapid(Water cooling)
6	Annealing	0 or 1	Temperature=900 °C, Timing: about 1 h, Cooling rate: rapid(Water cooling)
7	Aging (400 °C)	0 or 1	Timing: 1h<t<3h ,Cooling rate: Slow (air cooling)
8	Aging (500 °C)	0 or 1	Timing: 1h<t<3h ,Cooling rate: Slow (air cooling)
9	Aging (600 °C)	0 or 1	Timing: 1h<t<3h ,Cooling rate: Slow (air cooling)
10	Aging (700 °C)	0 or 1	Timing: 1h<t<3h ,Cooling rate: Slow (air cooling)
11	Aging (800 °C)	0 or 1	Timing: 1h<t<3h ,Cooling rate: Slow (air cooling)
12	Hot-rolled	0 or 1	Temperature: 800 °C, Timing: 6 min
13	Cold-rolled	0 or 1	
14	Extrusion	0 or 1	
15	Heat treatment (550 °C)	0 or 1	Timing: t <1h, Water cooling
16	Heat treatment (600 °C)	0 or 1	Timing: t <1h, Water cooling
17	Heat treatment (650 °C)	0 or 1	Timing: t <1h, Water cooling
18	Heat treatment (700 °C)	0 or 1	Timing: t <1h, Water cooling
19	Heat treatment (750 °C)	0 or 1	Timing: t <1h, Water cooling
20	Heat treatment (800 °C)	0 or 1	Timing: t <1h, Water cooling
No.	Outputs	Range	Details
1	Martensite Finish (Mf)	-140 to 644	
2	Martensite Start (Ms)	-140 to 644	
3	Austenite Start (As)	-140 to 644	
4	Austenite Finish (Af)	-140 to 644	
5	Martensite Peak (Mp)	-140 to 644	
6	Austenite Peak (Ap)	-140 to 644	

Table 2 Three validation cases; The features and the outputs are presented based on the various references. If a feature is 0 for all the cases it is eliminated from the table.

No.	Input features	Validation sets						
		Case 1	Case 2				Case 3	
1	Atomic percent of Ni	50.3	50.3				50.3	
2	Atomic percent of Hf	20	20				20	
4	Homogenization	1	1				1	
8	Aging (500 °C)	0	1				0	
9	Aging (600 °C)	0	0				1	
14	Extrusion	1	1				1	
No.	Outputs	Ref. 1. (Karakoc et al. (Ref 91))	Ref. 2 (Karaca et al. (Ref 70))	Ref. 1 (Karakoc et al. (Ref 91))	Ref. 2 (Karaca et al. (Ref 70)) 500 °C	Ref. 3 (Karaca et al. (Ref 70)) 550 °C	Ref. 1 (Karakoc et al. (Ref 91))	Ref. 2 (Karaca et al. (Ref 70))
1	Martensite Finish (Mf)	65	110	126	120	150	142	165
2	Martensite Start (Ms)	102	118	152	130	160	184	175
3	Austenite Start (As)	104	142	154	160	185	186	200
4	Austenite Finish (Af)	134	160	180	175	192	223	210
5	Martensite Peak (Mp)	83	115	139	125	155	163	170
6	Austenite Peak (Ap)	103	150	153	170	190	185	205

Figures:

Fig. 1 General approach followed for data-driven ML material design methods.

Fig. 2 Last step of optimum NN configuration selection; maximum, minimum, and average R^2 values for outputs features (Mf, Ms, As, Af, Ap and Mp) after 20 iterations for 30 and 23 to 27 neurons for the first and second hidden layers, respectively.

Fig. 3 The schematic of the NN model; Single input/Multi-output with 2 hidden layers with 30 and 25 neurons, respectively.

Fig. 4 Regression plots for (a) All the output data points as a united category, $R^2=0.922$ (b) Ms, $R^2=0.901$ (c) Mf, $R^2=0.912$ (d) Mp, $R^2=0.911$ (e) As, $R^2=0.908$ (f) Af, $R^2=0.900$ (g) Ap, $R^2=0.902$

Fig. 5 The histogram fit diagram; the abundance of the data points with a certain error. The Mean error and the RMSE are reported for (a) all data (b) Ms (c) Mf (d) Mp (e) As (f) Af (g) Ap

Fig. 6 Validation results; The comparison between the prediction and target results are presented for a) Case 1, b) Case 2 and C) Case 3. The red lines are the 10 predicted transformation temperatures.

Appendix A

NiTiHf Data Set; The description of the most important features, the number of data points and the related references.

Data set description	NO. of data sets	Reference
The study implemented the Suspended Droplet Alloying (SDA) fabrication method. This method showed to increase the TTs.	7	(Ref 83)
The work arc Melted the NiTiHf with additional post-processing like homogenization, extrusion, solutionizing, and several additional processes at different temperatures. The work demonstrates how each processing step changes the transformation temperatures.	47	(Ref 72)
The material Arc melted and homogenized before TT measurement with DSC instrument.	5	(Ref 92)
The study published in 1995 is focused on the ternary NiTiHf arc melted and homogenized. The TTs are measured for the different atomic percentages of NiTiHf and reported. Exploring the chemical composition effect on TTs is less studied which is the core of this work. A part of the data shows the effect of material heating with different temperatures on TTs for Ni ₄₉ Ti ₄₁ Hf ₁₀ . This study reported only the martensitic and austenitic peak temperatures. The rest of the outputs are assumed to be similar to the typically DSC results reported in this article. The data can be aggregated in the data set as the arc melting, homogenization and DSC measurements are the standard methods methods that exists even before the data of this publication.	24	(Ref 54)
The study is focused on Ni50.8Ti29.2Hf20 and Ni49.8Ti30.2Hf20 Materials. Material fabricated by induction skull melting followed by homogenization and extrusion. Then, the extruded wires were solutionized. After that, the materials were subjected to different heat treatments at different temperatures, such as heating, hot rolling, further solutionizing, and aging. Finally, the TTs were measured by the DSC technique for different conditions and reported in the article.	26	(Ref 69)
This study reports the additive manufacturing of Ni50.4Ti29.6Hf20 with a selective laser melting technique. Only one data set is added to the study.	1	(Ref 73)
In this study, the Ni50TiHf are studied with different amounts of Ti and Hf. The material is an electric arc melted and re-melted three times for homogenization., subsequent hot upsetting by 5–10% in a press, long-term annealing at 1073 K in argon, and quenching.	7	(Ref 85)
This study has a significant effect on the data sets as it presents the NiTiHf shape-memory alloy with low transformation temperatures. The study discusses that if the Hf amount is in the range lower than 12%, the NiTiHf can show very low transformation temperature, and the related data are added to the article. In this study, only the Martensitic finish transformation temperature is reported. The rest of the TTs are assumed based on the average difference of the TTs of the whole data set.	15	(Ref 58)
This study reported the TTs amounts for Ni50TiHf, Hf amounts of 6, 8, and 10 %. The materials are prepared with arc melting, followed by hot rolling, aging, or annealing, and the related TTs are plotted. The effect of the oxygen level is discussed for NiTiHf. The works (Ref 58) and (Ref 86) are presenting the TTs for Ni50.3Ti39.7Hf10 and Ni50Ti40Hf10 with hot rolling or without hot rolling, resulting in more than 100 °C differences in TTs.	12	(Ref 86)
The study implemented the melt spinning fabrication approach, followed by rapid quenching. Ni48.9TiHf with 5 different atomic percentages of Hf ranging from 8 to 20 percent are considered.	5	(Ref 84)

In this study, Ni50.3Ti29.7Hf20 was manufactured with induction melting following by homogenization at 1050 °C for 72 h and then extruded at 900 °C. The material then aged at different temperatures ranging from 300 °C to 900 °C, and the TTs reported. Results confirmed a continuous TT decrease for aging temperature 400 °C and a continuous TT increase for aging with 500 °C with increasing the aging time.	8	(Ref 70)
In this article, three different NiTiHf atomic percentages are investigated for the TT curves. The effect of thermal cycling is also investigated. Material heat treated up to 60 times at 800 °C or 900 °C. The study concluded that the TTs would become stable after 20 thermal cycles.	3	(Ref 60)
In this article, NiTi70Hf material with different atomic percentages of the Ni and Hf are fabricated by Arc Melting, and the related TTs are reported. The study is one of the rare ones which presents Hf atomic percentages lower than 10%.	3	(Ref 93)
The study measured the TTs for Ni38Ti50Hf12. This study considers the Ternary NiTiZr and the effect of thermal cycling on the TTs.	1	(Ref 94)
In this study, the Ni50.3Ti34.7Hf15 is fabricated by induction melting, homogenized at 1050 °C, hot extruded at 900 °C e (7:1), heat-treated at 900 °C for 1 h and then aged at various temperatures and times. This study discusses the effect of aging on the TTs.	5	(Ref 71)
In this work, Ni50.3Ti34.7Hf15 is fabricated through arc melting, homogenization, solution treatment, hot rolling, and aging at 450 °C or 550 °C at different timing from 0.5 to 72 hours. The data added to the model is the one related to 3h aging. It is observed that aging timing more than 10 didn't result in any significant changes in TTs.	3	(Ref 81)

Fig. 1 General approach followed for data-driven ML material design methods.

1653x717mm (38 x 38 DPI)

Fig. 2 Last step of optimum NN configuration selection; maximum, minimum, and average R^2 values for outputs features (Mf, Ms, As, Af, Ap and Mp) after 20 iterations for 30 and 23 to 27 neurons for the first and second hidden layers, respectively.

346x266mm (76 x 76 DPI)

Fig. 3 The schematic of the NN model; Single input/Multi-output with 2 hidden layers with 30 and 25 neurons, respectively.

414x227mm (87 x 87 DPI)

Fig. 4 Regression plots for (a) All the output data points as a united category, $R^2=0.922$ (b) Ms, $R^2=0.901$ (c) Mf, $R^2=0.912$ (d) Mp, $R^2=0.911$ (e) As, $R^2=0.908$ (f) Af, $R^2=0.900$ (g) Ap, $R^2=0.902$

798x279mm (76 x 76 DPI)

Fig. 5 The histogram fit diagram; the abundance of the data points with a certain error. The Mean error and the RMSE are reported for (a) all data (b) Ms (c) Mf (d) Mp (e) As (f) Af (g) Ap

482x272mm (76 x 76 DPI)

Fig. 6 Validation results; The comparison between the prediction and target results are presented for a) Case 1, b) Case 2 and C) Case 3. The red lines are the 10 predicted transformation temperatures.

417x213mm (87 x 87 DPI)