

Recent Advancements in Study of Effects of Nano/Micro Additives on Solid Propellants Combustion by Means of the Data Science Methods

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ABSTRACT

The efforts of Russian-Indian research team for application of the data science methods, in particular, artificial neural networks for development of the multi-factor computational models for studying effects of additive's properties on the solid rocket propellants combustion are presented. The possibilities of the artificial neural networks (ANN) application in the generalisation of the connections between the variables of combustion experiments as well as in forecasting of "new experimental results" are demonstrated. The effect of particle size of catalyst, oxidizer surface area and kinetic parameters like activation energy and heat release on the final ballistic property of AP-HTPB based propellant composition has been modelled using ANN methods. The validated ANN models can predict many unexplored regimes, like pressures, particle sizes of oxidiser, for which experimental data are not available. Some of the regularly measured kinetic parameters extracted from non-combustion conditions could be related to properties at combustion conditions. Results predicted are within desirable limits accepted in combustion conditions.

Keywords: Solid rocket propellant combustion; Nano-additives; Combustion characteristics; Data science; Artificial neural networks; Multi-factor computational models

NOMENCLATURE

E_a	Activation energy (KJ/mol)
P	Pressure (bar)
r	Burn rate (mm/s)
Al	Aluminium
HTPB	Hydroxyl-terminated polybutadiene (elastomer binder)
CTPB	Carboxyl terminated polybutadiene
SSA	Specific surface area (m ² /g)

1. INTRODUCTION

Combustion of solid composite propellants is an extensively studied area in the field of rocketry. Composite solid propellant essentially consists of ammonium perchlorate (AP) particles held together in a cured matrix of hydrocarbon binders. In order to enhance the ballistic properties such as burn rate, catalysts like iron oxide, copper chromite, etc., are added. The morphology and particle sizes of these additives play a critical role in deciding the final combustion characteristics. The interdependency of many of these parameters makes the combustion characteristics very complex. Empirical formulas have been derived for some dependences based on extensive experimental data. The most common representation of burn

rate dependence on pressure is Saint Robert's law (Vielle's law). Besides pressure many other parameters like particle size of oxidiser, O/F ratio and ambient temperature also influence the burn rate of solid propellant rocket. Some theoretical models like BDP do take into account the complexities of combustion zone marked with interlinked physicochemical processes in both gaseous and condensed phases. Since experiments are hazardous, expensive and time consuming, dependable models that can predict many of the critical parameters are always explored. There is no generalised multifactor model that allows the prediction of the burning rate and other decisive parameters of a new solid propellant mixture for defined ranges of pressure from the properties of the constituting additives like catalyst.

The burn rate of the solid propellants can be altered and tailored by the addition of catalysts. Transition metal oxide catalysts are added as catalysts for burn rate enhancement. It has been observed that there is a significant increase in the catalytic activity when the catalyst is present in nano size range. Recent years have witnessed synthesis of large number of nano catalysts as possible burn rate modifiers. However, relevant combustion studies in propellants using these additives are mostly confined to decomposition studies using conventional thermal analysis techniques. Lack of sufficient experimental data and complexities involved in any theoretical modelling calls for methods like artificial neural networks (ANN) which

can predict properties given a pattern of reliable test data. ANN can be an effective tool to relate the final combustion properties to many decisive parameters in a complex heterogeneous system like composite solid propellant.

In this work, we present results of ANN usage for creation of generalised models for combustion of AP-HTPB based composite solid propellants containing catalyst additives with different properties. The effect of few critical parameters such as particle size of catalyst, surface areas of oxidiser particles, kinetic parameters like activation energy and apparent heat release estimated from thermal analysis on the final ballistic property of AP-HTPB based propellant composition has been modelled using ANN methods. Previous attempts of using the ANN technique on propellant systems^{4,5} show the simplicity and feasibility of such an approach. The analytical platform (Loginom) used for creating and operating the model was developed by BaseGroup Labs⁶. The data used for training the model and validation were taken from either published literature⁷⁻¹⁶ or studies carried out at Energetic Materials Research Lab, IISc Bangalore.

2. ANN TECHNIQUE AND MODELLING

The ANN can be considered as a universal tool for multidimensional approximation. The ANN technique used here is the same as given in⁴. The details of the analytical platform (Loginom) can be seen at⁶. We have attempted three models: ANN model-1 (correlating the effect of specific surface area of catalyst and AP on burn rate), ANN model-2 (correlating the effect of apparent heat release on burn rate) and ANN model-3 (correlating the effect of activation energy on burn rate) at different operating pressures.

Figures 1(a), 1(b) and 1(c) shows the schemes of ANN model-1, model-2 and model-3 respectively. Model-1 consists of 1 input layer, 1 hidden layer and 1 output layer. The input layer consists of nine input neurons. The inputs considered are type of binder, AP % composition used in the propellant mixture, binder % composition in mixture, Al % composition in mixture, Al particle size, AP specific surface area, catalyst specific surface area, catalyst % composition in the mixture and operating pressure. The hidden layer consists of eight neurons and the output layer consists of one neuron, i.e., the burn rate. This model was trained using the data taken from⁷. The complete data set (training and validation) for ANN model 1 is given in Table S1 and Table S2 in the supplementary file. Among a total of 273 data points, 259 data points were used to train the model and 14 data points were used to validate the model. The data consists of 17 catalysed (with Fe_2O_3 as catalyst) formulations and 4 non catalysed formulations of AP-HTPB; 16 catalysed (with Fe_2O_3 catalyst) and 4 non catalysed formulations of AP-CTPB. The AP chosen in this study are a blend of different particle sizes (400 μm , 200 μm , 7-11 μm and 10 μm). This different particle size and its effects are indirectly incorporated into the model through the SSA of AP that is used as the one of the input parameter in the model.

Model-2 consists of one input layer of two neurons (Pressure and heat release), two hidden layers with 4 neurons and 3 neurons respectively and one output layer with one neuron (burn rate).

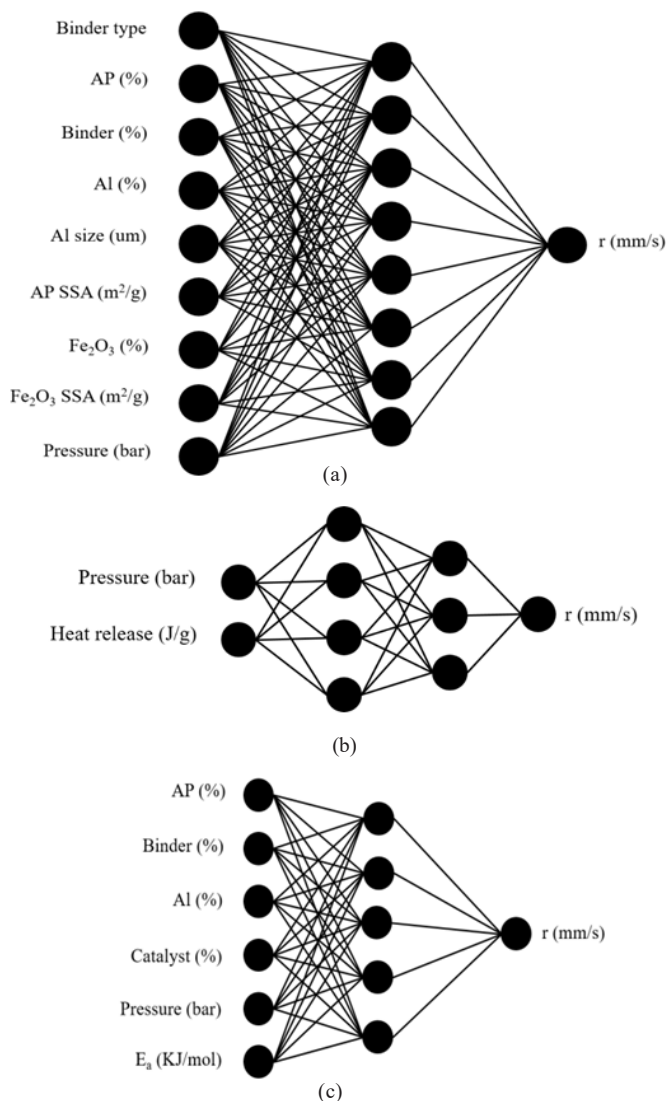


Figure 1. (a) Schematic of ANN model-1, (b) Schematic of ANN model-2, and (c) Schematic of ANN model-3.

Different operating pressures and heat release are given as input and the burn rate is obtained as the output. Data points used to train the model-2 were obtained by experiments carried out in-house and is given in Table 1. 12 data points were used to train the model and 3 data points were used to validate the model. An additional hidden layer is added to improve the accuracy. The relevant details are given in Table S3 and S4 in the supplementary file.

Model-3 consists of 1 input layer, 1 hidden layer and 1 output layer. The input layer consists of six neurons with inputs such as AP % composition in mixture, binder % composition in mixture, Al % composition in mixture, catalyst % composition in the mixture, operating pressure and activation energy. The hidden layer consists of 5 neurons and the output layer contains 1 neuron for burn rate. Model-3 was trained using 103 data points taken from¹⁰⁻¹⁶. The data points are drawn from experiments carried out using different catalysts. The details of the dataset (training and validation) is as shown in table S5 and S6 in the supplementary file. The primary input, namely activation energy used is independent of the catalyst. This

facilitates in establishing a direct relation between activation energy and burn rate.

In all three models, the training cycles were more than 10000 epochs and a sigmoid transfer function with slope steepness factor of 1 was used.

3. EXPERIMENTAL DETAILS

Most of the data used in the models are taken from reported literature. However, to relate the effect of energy release calculated from calorimetric techniques to burn rate irrespective of the propellant composition a small data set was compiled from burn rate experiments carried out at IISc using Crawford bomb technique. Three forms of manganese oxide and one form of iron oxide, all in nano size range, were used to study the effect of nano catalyst additive in AP-HTPB propellant combustion. The details of the propellant composition are as given in Table 1. Specific preparatory routes were adopted for preparing the nano catalyst. MnO_x E was prepared by a novel electrospinning technique developed in-house^{8,9}. Burn rate was measured at three different pressure values namely 30, 50 and 70 bars in N_2 atmosphere. The oxidiser and binders were obtained from VSSC Trivandrum. Apparent heat release of AP-HTPB-catalyst systems were estimated by a differential scanning calorimeter (Perkin Elmer DSC 8000) at ambient conditions. The samples (~ 10 mg) were taken in a hermetically sealed aluminium pan. An empty sealed aluminium pan was used as a reference. The heating cycle employed was a rate of 10 °C/min from 50 °C to 450 °C with N_2 as a purge gas.

Table 1. Heat release estimated from DSC studies for different propellant compositions

Propellant	Catalysts	Heat release in J/g	Pressure (bar)	Burn rate (mm/s)	Pressure index 'n'
AP-HTPB	No	440	30	6.19	0.4
			50	7.31	
			70	8.881	
AP-HTPB	Fe_2O_3	1757	30	12.22	0.46
			50	18.91	
			70	21.74	
AP-HTPB	MnO_x -A	873	30	5.6	0.43
			50	8.67	
			70	11.72	
AP-HTPB	MnO_x -B	1438.2	30	6.1	0.54
			50	9.6	
			70	13.04	
AP-HTPB	MnO_x -E	1893.9	30	10.05	0.8
			50	16.11	
			70	21.22	

4. RESULTS AND DISCUSSION

4.1 ANN Model 1

The trained ANN model-1 was validated by predicting the burn rate for a separate set of data points, which were not used in training set. This is as shown in Fig. 2(a) as a scatter

graph. The details of the validation are shown in table S2 in the supplementary file. It can be seen that ANN model-1 predicts burn rate with fairly good accuracy when compared with reported experimental values⁷. It is seen that the maximum % deviation of the ANN model predicted r with that of experimental r is less than 5 per cent and the model can now be used to bring about predictive correlation between the various input parameters and the burn rate. One such correlation is as shown in Figs. 2(b) and 2(c).

Figure 2(b) shows the ANN model-1 predicted burn rate variation with change in specific surface area of the catalyst used (in this case Fe_2O_3) at different operating pressures. All other input parameters were kept constant during modelling the relation between burn rate and SSA of catalyst (i.e., Binder = HTPB, AP = 76 %, HTPB = 12 %, Al = 10 %, Al size = 15 μ m, AP SSA = 2756 m^2/g , Fe_2O_3 = 2 %). It can be seen that at the three different pressures used for prediction, the burn rate increases with increase in catalyst SSA reaching a maximum value at a certain point and then decreasing. The increase in burn rate with SSA is expected, typical of any heterogeneous catalyst system wherein higher contact point facilitates higher reaction rates. The decrease after a peak predicted by ANN model could also be justified as higher surface energy associated with higher surface area possibly favours agglomeration and thereby negating the catalytic activity. The model can predict any possible SSA window wherein the burn rate gets either modified or remains unchanged. It can also project burn rate values at any pressures of interest without carrying out any experiment.

Figure 2 (c) shows the ANN model-1 predicted burn rate variation with change in SSA of AP at different operating pressures with other input parameters kept constant (i.e., binder = HTPB, AP = 76 %, HTPB = 12 %, Al = 10 %, Al size = 15 μ m, Fe_2O_3 SSA=3 m^2/g , Fe_2O_3 = 2 %). The trend shows that there is a burn rate increase with increase in SSA of AP. Increase in burn rate of AP based propellants with decrease in particle size of AP is well understood. Similar predictive comparisons can be done between different input variables with ease using the trained ANN model-1. The trend observed in this model appear similar to experimental data reported in the literature¹⁷ wherein effect of particle size of AP on AP-PS propellant burn rate was studied.

4.2 ANN Model 2

After the successful implementation of ANN technique to predict combustion characteristics in unexplored regimes, we made an attempt to relate burn rate to heat release obtained from thermal analysis technique, like differential scanning calorimetry (DSC) using a trained ANN model-2. It can be seen from Fig. 3(a) that the catalyst enhances the burn rate and is specific to the nature of catalyst. Besides the effect of pressure is more pronounced in catalysed systems.

The training set is shown in Fig. S2 as a scatter graph in the supplementary file. The % deviation of the ANN predicted burn rate from that of experimental values were less than 5 %. Given only the heat release obtained from a calorimetric technique like DSC, burn rate can actually be predicted within this accuracy. This has been validated as shown in Fig. 3(b).

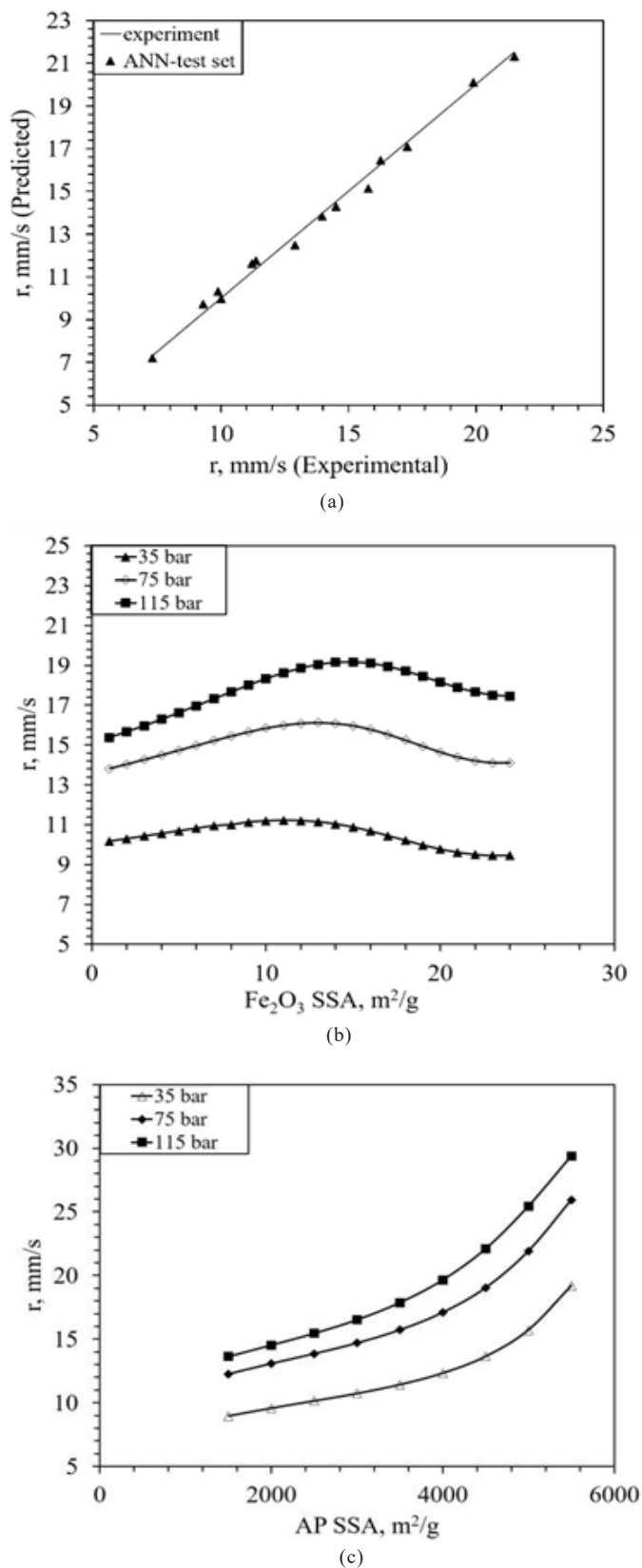


Figure 2. (a) Burn rate predicted by ANN model-1 vs experimental burn rate, (b) Effect of catalyst specific surface area (SSA) on burn rate at different operating pressure conditions predicted by ANN model-1, and (c) Effect of AP specific surface area (SSA) on burn rate at different operating pressure conditions predicted by ANN model-1.

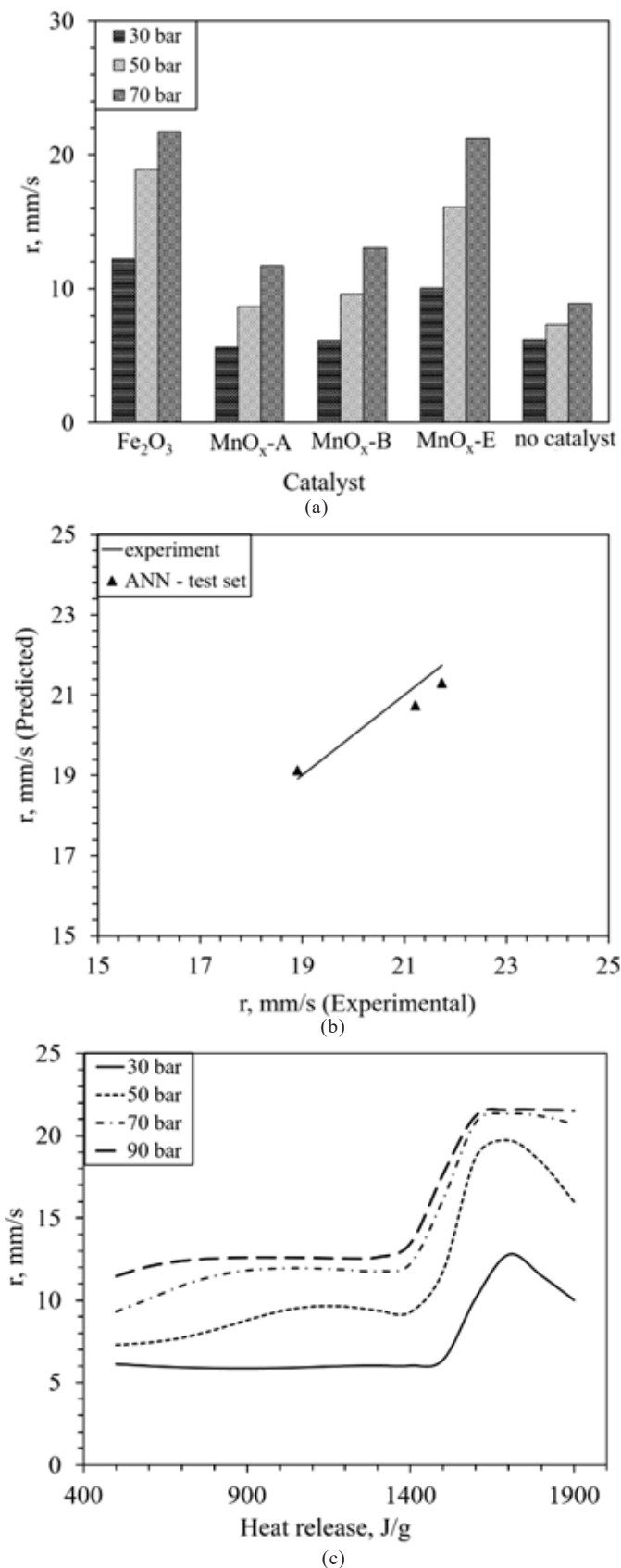


Figure 3. (a) Effect of different nano catalysts on burn rate at different operating pressure conditions, (b) Burn rate predicted by ANN model-2 vs experimental burn rate, and (c). Burn rate vs Heat release at different operating pressures predicted by ANN model-2.

Further extrapolation into regions of different pressures and energy values are possible using this model as demonstrated in Fig. 3(c). This is important as any new catalyst reported may not always be tested in a propellant composition and claim as a possible burn rate modifier is based only on the observed enhanced heat release obtained from thermal studies. While the increase in burn rate with heat release can be understood, the inflection beyond a certain energy level predicted in the present case need substantial backing from extensive experiments, primarily because no such observations have been made hitherto.

4.3 ANN Model 3

Yet another relation that we could establish is between activation energy and burn rate which is given by the ANN model-3. Reported activation energy normally measured from thermal analysis techniques available in the literature have been used in this model. No such relation is reported in the literature as per our understanding. The trained ANN model-3 was validated by predicting the burn rate for a separate set of data points taken from¹⁰⁻¹⁶, which were not used in training set. This is shown in Fig. 4 (a) as a scatter graph. The details of the validation are as shown in table S6 in the supplementary file. It can be seen that the ANN model-3 predicts burn rate with very good accuracy. The maximum % deviation of the ANN model predicted r with that of experimental r is less than 10 % and the ANN model can now be used to bring about predictive correlation between the various input parameters and the burn rate. Such a correlation is as shown in Fig. 4 (b). It shows the ANN model-3 predicted burn rate variation with change in activation energy of the composite solid propellant with different catalyst at different operating pressures. The other input parameters were kept constant (AP = 67.1 %, HTPB = 13.9 %, Al = 18 %, catalyst = 1 %). It can be seen at the two different pressures used for prediction that the burn rate decreases with increase in activation energy reaching a minimum value at a certain point and then there is no change further. This also augurs a critical value of activation energy independent of pressure. Similar correlation can be done between other input variables and burn rate to study their effects. This becomes more interesting when the values used are for different catalytic system and model can predict the burn rate independent of its chemical constitution. The kinetic data generated from thermal analysis are plenty in literature for different propellant composition and are never extrapolated to final ballistic properties like burn rate. This relation is another interesting outcome of this model.

5. CONCLUSIONS

In this paper, we present the results of usage of artificial neural network (ANN) technologies to relate certain combustion parameters like burn rate with few physicochemical properties of constituting components in heterogeneous systems like composite solid propellants. Though the complexities of the combustion zone in such systems are difficult to model easily, ANN can effectively

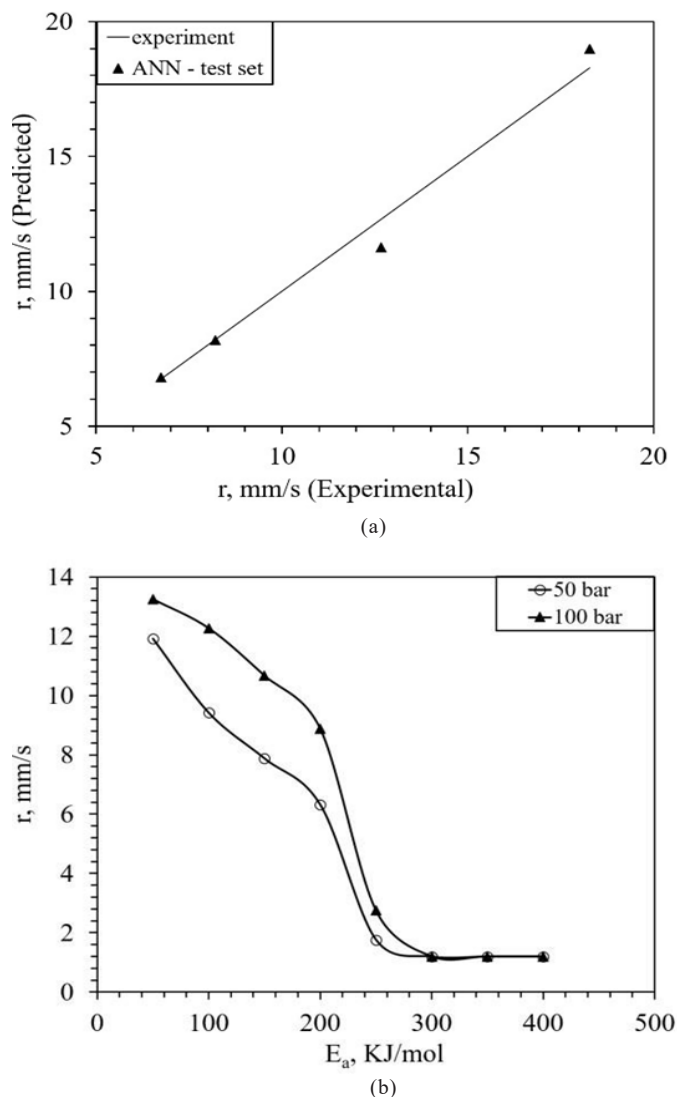


Figure 4. (a) Burn rate predicted by ANN model-3 vs experimental burn rate and (b) Effect of activation energy on burn rate at different operating pressure conditions predicted by ANN model-3.

predict a property as long as the pattern used for training is reliable and reproducible. The three models exhibit good predictive capability to show the dependence of burn rate on specific surface area of catalyst and AP, apparent heat release and activation energy obtained from thermal analysis measurements at different operating pressures. Such models can be considered as the foundation for further development of models through ANN for predicting the effects of more combustion parameters in a fast and relatively easy way. This approach can be extended further by creating an integrated model which combines all the parameters of the three models. Such a complete model will be able to take into account many of the characteristics of propellant composition along with its thermodynamic properties to give a very good prediction of burn rate. Also an inverse problem approach can be done which will be able to predict propellant compositions with specific physical and chemical properties to give a particular burn rate.

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ADDITIONAL INFORMATION

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