Prediction of Mechanical Properties of Plasma Sprayed Thermal Barrier Coatings (TBCs) with Genetic Programming (GP)

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Abstract—*The mechanical properties especially hardness and porosity of plasma sprayed thermal barrier coating (TBC) play a major role in deciding their lifetime and performance with respect to input process parameters such as power input of plasma jet, coating thickness, stand-off distance and type of coating. Sources of mechanical properties values are experimental measurements only, and empirical correlations are to be built up (without appropriate fitting techniques), however, these are often too complicated, expensive and time consuming and can lead to erroneous results. Genetic programming (GP) is the most common approach from various evolutionary computation methods using multivariate regression fitting for the modelling of various systems. This study presents a new model for estimating the mechanical properties of TBC using GP. On the basis of a training data set, different genetic models for mechanical properties with great accuracy were obtained during simulated evolution. The newly developed GP-based computational model provides a more accurate prediction of mechanical properties compared to the empirical correlations, and the results can then be utilized to estimate a future set of parameters based on the historical data.*

Keywords—*Hardness, Porosity, Thermal Barrier Coatings, Plasma Spraying, Genetic Programming.*

I. Introduction

Thermal barrier coatings (TBCs) are attracting ever-increasing attention in relation to various industrial, automobiles, gas turbines, power generators, and aerospace applications as they prepare a suitable engineering surface for existing materials and alloys due to their ability to withstand severe working conditions such as high temperature, oxidation, excessive wear and so on [1] [2]. Generally, TBCs provide thermal insulation, oxidation, wear, corrosion, chemical degradation resistance by coating the surface with industrial ceramic coatings using an atmospheric plasma spray coating method [3]. Moreover, TBCs are ceramic coatings applied on the metal substrate in order to increase the life of engineering materials. Furthermore, TBCs are formed by two layers namely, a metallic bond-coat layer, which prevents oxidation and corrosion and increases adhesion strength [4] and a ceramic coating layer, which protects metal from severe working conditions [5] [6] [7][8]. Plasma-sprayed Alumina, Alumina, and Titania, Zirconia-based ceramics (Partially Stabilized Zirconia and Super-Z alloy) are the most significant coating materials because of their low thermal conductivity, excessive hardness, high wear resistance, oxidation, and reduced porosity as a result of the plasma spraying process [7] [9]. However, the durability of TBCs under severe mechanical loading conditions encountered remains one of the main issues outstanding. Hence, the development of TBCs demands better understanding of the mechanical performance of the coating materials under different process parameters to ensure the life and reliability of the various components.

During the past one to two decades, various plasma-sprayed TBC systems have been developed and characterized to determine their mechanical properties at ambient as well as at elevated temperatures. These attempts have been made using TBC substrate systems. However, various investigations have been carried out to present a multitude of mechanical properties of different TBC materials which can be conveniently employed as a history of past data. Mechanical properties of plasma-sprayed TBCs have been evaluated and compiled in this report to provide them as a history of design database. The mechanical properties include hardness and porosity determined under various input conditions. TBC associated with porosity so that they would be expected to reveal some of mechanical properties. The directionality effect has been quantified through hardness measurements and is also presented in this report [10] [11] [12].

The four different commercial, industrial ceramic materials are used in hardness and porosity testing at ambient temperature (25°C) for test specimens and spray parameters to plasma-spray. The final surface finish of test samples was achieved using a 500-graded diamond grinding wheel [13] [14].

A. The Atmospheric Plasma Spraying Process

This uses high temperature plasma due to ionization of gas produced from the high strength electric arc which is struck between the cathode (tungsten electrode) and an anode (nozzle) in the presence of the mixture of Argon and nitrogen/hydrogen in the chamber. Coating material particles are heated within the plasma jet, and at high velocities, molten droplets sprayed onto the surface of a substrate to produce the coating. APS ceramic coatings are widely employed in those engineering applications which demand wear resistance, corrosion resistance and high strength at elevated temperatures [15][16].

B. Genetic Programming (GP)

Genetic Programming (GP) is an estimation or automated machine learning method instigated by natural evolution like biological growth to develop computer programs with high fitness to a particular process output by transporting a population of small specific programs [17]. GP is used for regression and binary classification problems. Various programs are produced by mutation and crossover using computational analog. We assign to the programs created classifiers. The suitability of each variable is evaluated using a fitness function to select the successful variables. GP also uses the principle of Darwinian Natural Selection to select, evolve and reproduce "fitter" programs better than would a random search process. The symbolic regression function is too complex to assess by human trial and error functions whereas the machine learning technique of GP maps a set of inputs data to known output data of engineering problems to determine data mining and knowledge discovery. GP thus provides a significant benefit in many areas of science and industry [18].

The solutions generated by GP are computer programs which are easy to inspect, evaluate, test and are also easy to understand in terms of the relationship between input variables and output data as well as to tap the uncovered relationships that were unknown before. GP output results are fed into both the model and the constants concurrently. It is very successful in solving a broad range of problems involving systems modelling (SM), curve fitting (CF), data modelling (DM) and symbolic regression (SR). Applications include industrial process control (IPC), financial trading (FT), time series prediction (TSP), economic modelling (EM), optimisation and scheduling (O & S), medicine, signal processing (SP), entertainment and computer games (E & CG) [19] [20] [21].

Symbolic regression discovers both the working model of a target function and its fixed coefficients, or at least an approximation to these and differs from other types of regression e.g. polynomial regression which confines itself to merely trying to determine the coefficients of a pre-defined order of a polynomial. The GP technique involves obtaining evolutionary algorithms (EA) based on highlights of the study of natural selection and evolution to solve problems using a process of first generating many random problem solvers (programs) rather than focussing on explicit design and analysis. Each program is executed and rated according to a fitness value defined by the developer similar to biological evolution in nature. EA selects the best programs in each generation and produces them [22][23].

This paper presents a new model for estimating/predicting the mechanical properties i.e., porosity (%), and hardness (RHc) of TBCs with various input parameters required for characterization using a genetic programming approach. In addition to this, it includes deriving empirical correlations used in forecasting mechanical property values to an accuracy level of 98.99% for characterization of MP of TBCs using Discipulus^{TM} software. The new model was designed to be simpler as it eliminates the numerous computations involved in any equation of state applications. These equations would help in developing and testing the developed or would be developed correlations and empirical relations in the future [24].

II. Materials and Methods

The substrate made of mild steel and standard dimensions 48 mm × 48 mm × 6 mm was selected. One of the substrate flat sides was initially cleaned with acetone, subjected to grit blasting (using alumina) and degreased in ethyl alcohol. Several jigs and fixtures were used for mounting the specimens according to the spraying requirements. These substrates were secured tightly in suitable holders after cleaning. The initial thickness of the substrate was measured on a few spots to measure the bond coat and the ceramic coating thickness. The substrate on the mount was finally degreased again with ethyl alcohol. These steps were followed very precisely because any quantity of contamination on the substrate was likely to provide a weak point of adhesion for the subsequent coating. In the present study, 96% high-purity of Alumina Ceramic (Al_2O_3), Alumina Titania (Al_2O_3+TiO_2), Partially Stabilized Zirconia ZrO_2 (PSZ) and Super-Z alloy (20 % of Al_2O_3 + 80% of PSZ) is used as the workpiece material[5] [6] [7].

A bond coat of 50 to 100 µm thick commercial nickel based alloy Ni33CrAlI, Amdry 962 (referred to as Nickel Chromium Aluminum Yttrium, NiCrAlY) powder was spray coated onto the substrate using a plasma spray technique with pressure at 60 kW, SULZER METCO spray systems. The desired number of passes of the plasma gun over the substrate was calculated based on the required thickness, typically the system was set to deliver a 50 µm thick coating per pass and was present in the computer controlled plasma spray system. The thicknesses of the substrate plus the bond coat were measured at the same spots where substrate thicknesses had been measured earlier and the average bond coat thickness was ascertained. The substrate was kept air cooled during spraying, the oven-dried plasma sprayable TBC powders of the desired composition were then plasma sprayed onto the bond-coated substrates. Here again, the number of passes required for deposition of a thickness of coating was ascertained by measuring the thickness of the ceramic coating after a single pass under the present chosen conditions of spraying. The thickness of the coated substrate was measured (metallic bond-coat layer and ceramic coating layer) on the substrate [24].

A. Porosity of TBC

The porosity of TBC prepared by thermal spraying technique is an important aspect in deciding various strengths and insulation properties such as the process of welding. Porosity referred to as the void fraction (a measure of the void or space) of a coated material. It is typically characterized as a percentage (0 to 100%) of the voids volume within the total volume. Pores (i.e., their size, shape and amount) reduce the strength of ceramics because they reduce the cross-sectional areas over which a load can be applied and, consequently, lower the stress that these materials can support. It can take on various forms such as open, closed, connected, elongated, etc. Porosity is a general occurring feature of thermal spray processes and is a very dynamic process, involving thermal, kinetic and chemical processes with serious issues as regards porosity in TBCs. The measurement of porosity is easy to understand but difficult to carry out. Numerous techniques are used to estimate porosity and the light microscope image analysis is the most well-known method which measures porosity along with checking the thickness, interfaces, contamination, and so on. Preparation of the sample includes sectioning, cleaning, mounting, grinding and polishing before microscopic inspection.

ASTM E2109-01 comprises methods to give porosity grades on metallographic samples and are prepared according to ASTM E1920 [25]. It uses an area measurement where area is equal to the porosity of the volume as long as the small pores are evenly dispersed [19] [26].

III. GENETIC PROGRAMMING METHODOLOGY

Data groups of experiments considered for the analysis of mechanical properties of different types of TBCs having spray factors such as standoff distance (100-140 mm), input power (16-40 kW) and thickness of coating (100-300 µm) were taken. The data groups were randomized using Discipulus™ software. The randomized data groups were provided to the software in three groups viz., training, validation and applied [17]. Trial runs were carried out to find out the best parameters that generated an optimal solution in the minimum possible time. Initially, the runs were conducted with the default one by one the parameters such as population size, crossover rate, DSS subset size, and the FPU registers used were varied to find optimum values [18]. The trials showed the following results. The population size of 600 was optimum rather than the default setting of 500. A higher crossover rate (75% non-homologous and 25% homologous) was found to be optimum. A smaller DSS subset size 60 was more optimal than the default 100. The above factors favourably affected result generation.

A. Regression

Regression analysis is a statistical method and symbolic regression discovers both the working model of a target function and its fixed coefficients, or at least an approximation to these.

B. Fitness Measurement

Fitness is nothing but how far the data value predicted by the GP coincides with the experimental value.

C. Correlation Coefficient, r/R

The quantity refers to linear correlation coefficient to measure the strength and direction of a linear relationship between two variables. The value of r lies such that $-1 < r < +1$ [27]. The (+) and (–) signs are utilized for positive and negative linear correlations, respectively. If *P* and *Q* have a strong positive linear correlation, r is close to +1 and indicates a perfect concrete fit i.e. a relationship between *P* and *Q* such that as values for *P* increase, those for *Q* also increase. Whereas, if *P* and *Q* have a strong negative linear correlation, r is close to -1 indicating a perfect negative fit showing a relationship between *P* and *Q* such that as values for *P* increase, those for *Q* also decrease. A value $r = 0$ for no linear/ weak correlation and a value near zero indicates a random, nonlinear relationship between the two variables (inputs and output). The square of the correlation coefficient gives the coefficient of determination, r^2, to find the proportion of the variance of output that is predictable from the inputs. It helps us to determine how certain one can be in making predictions from a defined model. r^2 is defined from the ratio of the illustrated variation to the total variation in the range of $0 < r^2 < 1$ signifies the strength of the linear correlation between *P* and *Q* or represents the percentage of the data which is closest to the line of best fit [28]. If $r = 0.977$, then $r^2 = 0.994$, which means that 99.4% of the total variation in *Q* can be explained by the linear relationship between *P* and *Q* and the remaining 0.6% of the variation in *Q* continues unexplained [26].

D. Factors involved in GP Modelling

The various parameters involved in modelling GP are tabulated in Table I. It shows the flow of set required to achieve the final model which could provide you with a mathematical model satisfying the above conditions of quantity involved.

TABLE I

VARIOUS FACTORS INVOLVED IN MODELLING GP

Terminal Set	T = {P, Random-Constants}
Instructional Set	F = {Arithmetic, Subtraction, Division, Addition, Multiplication, Exponential and Trigonometric}
Fitness r^2	The square root of the sum of the square of absolute value of the differences (errors), between the program's output and the observed data.
Termination	An individual emerges whose sum of absolute errors is less than specified
Parameters	(a) Required number of runs are completed, or (b) Required correlation coefficient is obtained

IV. RESULTS AND DISCUSSIONS

In GP modelling, it is important select proper instructions from set *F* and available terminal genes from set *f*(0) [17]. From these, the evolutionary process will try to build an organism (i.e. mathematical model) as fit as possible for the prediction of mechanical properties and which consists of both instructions and function genes behaving similarly to the nature of computer programs which differ in form and size [28] [29]. Measurement acquired converted into three independent data sets: training, validation, applied data sets. type of coating, coating thickness, stand-off distance, input jet power was used as independent input variables and the porosity and hardness number as the dependent output variable. From the training data set, different models for mechanical properties were developed by the genetic programming [22] [23][30]. Using GP simulation, the best mathematical model for porosity and hardness is given by Eq. (1) and Eq. (2), respectively.

$$\boldsymbol{Porosity} = 0.03\left(\sin 2\left((-2+4\times TC)2-1\right)-1-T\right)+0.74(P+SD+18J+7TC-13+4I) + 4|\sin(\cos(2\sin(H-1.92)-SD)/I)| \tag{1}$$

Where;

$\boldsymbol{A} = 0.03\cos(2.2-P)+0.5/(TC+P)$

$\boldsymbol{B} = \cos(2.2-P)\times 3.3|\sin A|\sqrt{\left((6.6|\sin A|/P)+1.25\right)}$

$\boldsymbol{C} = B\sin^2 T+\sin^4 T$

$\boldsymbol{D} = 8\cos^3 4C^2/P^3-\{2\cos 4C^2/P\}$

$\boldsymbol{E} = B\sin^2 T+\sin^4 T-D^3-1$

$\boldsymbol{F} = E/\{(D^6-2D^3+1)\sin(1-P)\}+\cos(1.5+0.75SD)$

$\boldsymbol{G} = |9.1\sin(3.1SD\times\cos^2(1.5+0.75SD))|$

$\boldsymbol{I} = 6.6|\sin A|/P+\sin^2 T+1-\{\cos^2(1.5+0.75SD)\}+3.1\cos^2(1.5+0.75SD)/(1.6\cos^2 T+1.1)$

$\boldsymbol{H} = (1.6\cos^2 T)+1.1+F\sin(2G/P-1.36)^2+2\cos^2 T$

$\boldsymbol{J} = F\sin(2G/P-1.36)^2+2\cos^2 T+\sin^2((-2+4TC)^2-1)-1$

$$\boldsymbol{Hardness} = 4\cos(3.16\cos P)+(3.16\cos(0.52P+0.52SD+0.52TC)+2.32)+M + 2\sin(2\sin 2N+M)/2.234+\cos TC+SD-0.52P + 2\cos(\cos(TC+SD-0.52P)+1.43+P) \tag{2}$$

Where;

$\boldsymbol{A} = (P(\cos(P+1.21)+TC)-1.91)/PT$

$\boldsymbol{B} = 2\left((P(TC+\cos(P+1.21))-3.82)/PT\right)-TC/T+(P(\cos(P+1.21)+TC)+1.91)/PT^2$

$\boldsymbol{C} = \sqrt{(0.95/B)+TB}$

$\boldsymbol{D} = \cos C+B/P-2B$

$\boldsymbol{E} = \cos(2(D+B+T)^2+D)/(-0.43)$

$\boldsymbol{F} = \left\{\cos\left(\sin\left(E^2/(E^2-1)\right)DE^2/E^2-1\right)+(DE^2/E^2-1)/(-0.65)\right\}^4-0.95+TC$

$\boldsymbol{G} = |(\cos^2 F-1)/(0.03-2)(DE^2/E^2-1)\{\cos(\sin(E^2-1))(DE^2/E^2-1)+(DE^2/E^2-1)/-0.65\}^4 /2T|$

$\boldsymbol{H} = \left(\left((0.11TG)-1.36\right)/(-0.73)-\left((DE^2/E^2-1)+G\right)/P\right)^2-1$

$$\boldsymbol{I^2} = \sin\left(\cos\left(\sin\left(2\left(\left(\left|(\cos((H/TC-1.36)SD))P\right|/T\right)-0.61\right)1.26\right)+(DE^2/E^2-1) + G/\sqrt{(H+1)-H}\right)^2\right)+1.74-(DE^2/E^2-1)+\left(G/\sqrt{(H+1)-H}\right)(P/(-0.97)/T)^2$$

$$\boldsymbol{J} = (\cos(\sin(2I)))^2-1+(DE^2/E^2-1)+G/\sqrt{(H+1)-H+I^2}+(DE^2/E^2-1) + G/\sqrt{(H+1)-H+I^2}/(\cos(\sin(2I)))^2-1+(DE^2/E^2-1) + G/\sqrt{(H+1)-H+I^2}$$

$$\boldsymbol{K} = (DE^2/E^2 - 1) + G/\sqrt{(H+1) - H + I^2}/(\cos(\sin(2I + P)))^2 - 1 + (DE^2/E^2 - 1) + G/\sqrt{(H+1) - H + I^2}\,/J$$

$$\boldsymbol{L} = \big((\cos(2\sin J)) + 0.002\big) + (DE^2/E^2 - 1) + G/\sqrt{(H+1) - H + I^2}/(\cos(\sin(2I + P)))^2 - 1 + (DE^2/E^2 - 1) + G/\sqrt{(H+1) - H + I^2}\,/J - 0.13/(0.422)$$

$$\boldsymbol{M} = K + L + 2\cos\big((TC(L + TC + 1.64)/T) - 0.033\big)$$

$$\boldsymbol{N} = 4\cos^4\big((TC(L + TC + 1.64)/T) - 0.033\big) - K + L + 2\cos\big((TC(L + TC + 1.64)/T) - 0.033\big)$$

Comparison between the experimental outputs and predicted outputs using the derived mathematical model obtained from GP are illustrated in Tables II and III for porosity and hardness of the TBC mechanical properties. Errors are very low and percentage of error is less than +/ -1% which shows that results are highly acceptable with this predicted model. Besides, Figures 1 and 2 show the regression fit for the % of porosity and hardness of the TBC mechanical properties.

TABLE II

COMPARISON BETWEEN EXPERIMENTAL AND GP VALUES OF POROSITY

No.	Type of Coating (TC)	Stand-off Distance (SD), mm	Thickness of Coating (T), µm	Input Jet Spray Power (P), KW	Porosity Output		Error %
					experimental	GP	
1	3	140	200	40	7.8	7.74	0.06
2	1	140	100	40	5.5	5.41	0.09
3	3	120	150	16	7.1	7.14	-0.04
4	2	100	100	25	7.05	7.01	0.04
5	3	140	100	30	4.85	4.9	-0.05
6	4	110	300	16	10	9.92	0.08
7	2	110	100	40	4.8	4.84	-0.04
8	3	110	150	25	6.4	6.44	-0.04
9	3	120	200	16	8.5	8.57	-0.07
10	2	140	100	40	5	5.1	-0.1
11	1	140	150	30	5.5	5.53	-0.03
12	3	100	100	40	4.1	4.09	0.01
13	1	100	200	16	11.3	11.28	0.02
14	4	140	100	30	4.4	4.35	0.05
15	1	100	100	25	7.4	7.47	-0.07
16	4	140	100	25	6.4	6.36	0.04
17	4	110	300	30	5.2	5.17	0.03
18	2	100	100	16	7.5	7.51	-0.01
19	2	120	150	25	6.3	6.39	-0.09
20	2	120	150	30	4.6	4.66	-0.06
21	2	140	150	25	6.9	6.95	-0.05
22	4	100	150	30	4.1	4.18	-0.08
23	4	100	100	25	6.1	6.15	-0.05
24	2	140	100	25	7.2	7.23	-0.03
25	3	110	300	25	6.7	6.61	0.09
26	2	100	300	16	11.1	11.17	-0.07
27	4	120	150	16	6.5	6.59	-0.09
28	4	120	150	30	3.85	3.846	0.004
29	4	100	200	30	4.3	4.32	-0.02
30	2	110	150	25	6.85	6.82	0.03
31	3	140	100	40	4.45	4.46	-0.01
32	2	120	300	25	6.8	6.86	-0.06
33	1	120	300	30	6.3	6.35	-0.05
34	2	110	300	25	7.2	7.23	-0.03
35	1	100	150	25	7.25	7.18	0.07
36	1	120	300	25	7.3	7.36	-0.06
37	2	140	150	16	9.4	9.39	0.01
38	4	100	200	16	8.6	8.59	0.01
39	4	110	100	40	3.95	3.948	0.002
40	2	120	300	30	5.85	5.86	-0.01
41	3	120	100	40	3.75	3.76	-0.01
42	3	100	100	16	7.1	7.03	0.07
43	3	140	100	25	6.9	6.83	0.07
44	3	120	300	25	6.4	6.34	0.06
45	1	140	300	40	12	11.99	0.01
46	3	140	150	25	6.5	6.49	0.01
47	3	110	150	16	7.6	7.56	0.04
48	3	100	150	30	4.6	4.69	-0.09
49	4	100	100	16	6.4	6.43	-0.03
50	2	100	300	30	6.2	6.12	0.08

51	1	100	100	40	5	5.02	-0.02
52	1	110	100	16	8	8.05	-0.05
53	3	140	300	30	5.85	5.9	-0.05
54	1	100	300	25	7.9	7.85	0.05
55	4	100	100	30	3.8	3.89	-0.09

TABLE III

COMPARISON BETWEEN EXPERIMENTAL AND GP VALUES OF HARDNESS

No.	Type of Coating (TC)	Stand-off Distance (SD), mm	Thickness of Coating (T), μm	Input Jet Spray Power (P), KW	Hardness Output		Error %
					Experimental	GP	
1	1	120	200	40	125.538	125.798	-0.259
2	2	110	150	40	119.502	119.734	-0.232
3	2	120	100	16	114.842	115.049	-0.207
4	3	120	200	25	128.622	128.342	0.280
5	2	140	200	40	100.721	100.848	-0.126
6	2	100	100	16	97.2315	96.9231	0.308
7	3	120	150	16	130.106	129.96	0.146
8	4	120	150	25	131.702	132.027	-0.324
9	2	110	150	16	92.5798	92.573	0.006
10	2	100	200	16	76.9135	76.8976	0.015
11	2	120	150	40	126.046	125.78	0.2653
12	4	120	300	16	106.873	106.544	0.329
13	4	140	300	16	84.2797	84.398	-0.118
14	3	140	300	40	101.512	101.206	0.306
15	1	120	100	16	123.462	123.654	-0.192
16	3	120	150	25	134.901	135.083	-0.182
17	1	140	200	16	88.4932	88.5517	-0.058
18	4	110	300	40	104.388	104.05	0.337
19	2	120	300	16	88.215	87.9218	0.29
20	2	140	200	16	78.276	78.0044	0.271
21	2	140	150	25	109.998	109.934	0.064
22	1	100	200	40	112.096	112.082	0.013
23	3	120	300	30	144.353	144.596	-0.242
24	1	110	150	16	101.513	101.621	-0.108
25	3	100	100	16	119.16	118.977	0.182
26	3	120	100	16	137.259	137.138	0.121
27	2	110	150	25	105.871	105.816	0.054
28	4	140	300	40	98.0383	98.0714	-0.033
29	4	120	150	16	127.112	127.006	0.106
30	1	100	200	16	87.0563	87.2872	-0.230
31	4	110	100	25	132.614	132.3	0.313

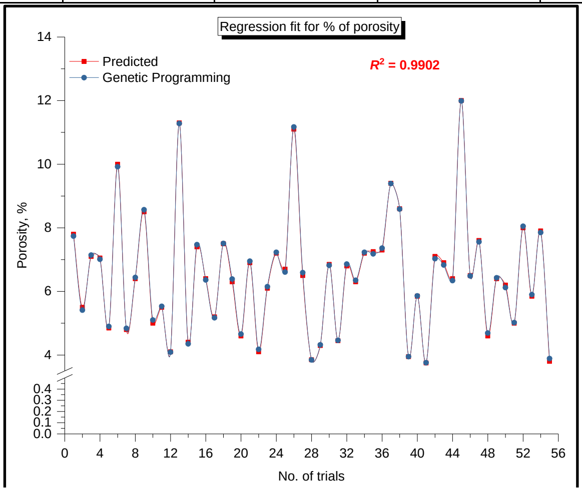

Fig. 1: Regression fit for the percentage of porosity

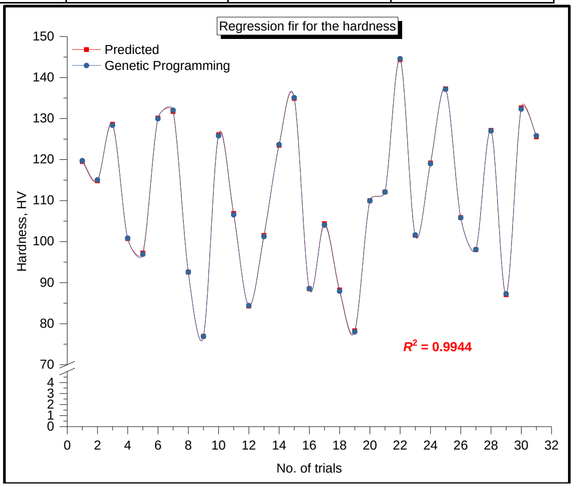

Fig.2: Regression fit for the hardness

V. CONCLUSIONS

In this paper, new models of the porosity and hardness of mechanical properties at different spraying parameters for various thermal barrier coatings were developed using GP. With the help of a computational model, the mechanical properties for TBCs involving various spray parameters by easy substitution without carrying out any experiments can be predicted. The comparison between the new GP-based model and the experimental results indicated that the new model is more accurate close to +/- 0.006 to 0.009. Therefore, the new model can be considered an alternative method to estimate the mechanical properties when the experimental measurement or correlations are not available. The correctness of solutions achieved by GP depends on correlated evolutionary parameters, the number of experimental results and their level of accuracy. To improve the structure of the model during evolution, more information supplied by providing the number of measurements and in the present proposed mathematical model for verifying the experimental results is subject to adaptation with reliability of about 99.4%. In the testing stage, the GP model gives the same result as found out during the experiment with the reliability of cent percent. The GP approach has thus proved to be a highly skilled and advantageous tool for recognizing correlations in data when no proper theoretical or other methods are possible or available.

CONFLICTS OF INTEREST

The authors have no conflicts of interest.

FUNDING

The authors received no financial support for the research and/or for the publication of this article.

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