Response Surface Methodology for Optimization of Plasma Spraying

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Response surface methodology was used to describe empirical relationships among three principal independent variables that control the plasma spraying process. The torch-substrate distance, the amount of hydrogen in the primary gas (argon), and the powder feed rate were studied. A number of dependent variables (responses) were determined, including the deposited layer roughness, density, hardness, chemical composition, and erosion rate. The technique facilitates mapping of the responses within a limited experimental region without much prior knowledge of the process mechanisms. The maps allow process optimization and selection of operating conditions to achieve the desired specifications of the plasma sprayed coating. To illustrate the approach, a simple system of WC-12%Co was deposited on a mild steel substrate. The resulting response surfaces were used to define optimum, or "robust," deposition parameters.

1. Introduction

DESPITE increased interest in the fundamentals of plasma spraying, there is still a lack of reliable models that relate engineering properties of coatings, such as hardness or density, to variations in process parameters or deposition geometry. This gap between the need to understand a process to optimize it and a growing demand for good plasma sprayed coatings can be filled temporarily by empirical modeling techniques. Response surface methodology (RSM) is a powerful empirical modeling tool to describe smooth relationships, for which physical underlaying mechanisms are generally not known or are not understood. The approach is based on formulation of a low degree polynomial, relating M observations Y_j ($j = 1 \ldots M$; here, $M = 5$) to N process parameters X_i ($i = 1 \ldots N$; here, $N = 3$). Usually, the cubic polynomial is the highest degree considered, and the lower the degree, the higher confidence in the model.

Transformation of process parameters X_i and/or responses Y_j (for example, through compression of the scale by log $[X_i]$, log $[Y_j]$) frequently helps to obtain a "best fit" between an experiment and a simple empirical model of low degree. A well-known example of this procedure is a single-variable least-squares curve fitting.

The response surface methodology is based on similar least-squares procedures, with similar results. The parameters of the equation that best fits the experimental results are obtained and assessed against the statistical significance. The number of variables usually involved in the response surface methodology is larger than one, and therefore, graphical representation of the response surfaces $Y(X_i)$ could be complicated. The predicted response surfaces Y (or contours of constant value $Y = Y_c$) can be mapped conveniently within the limited area of interest, if any two independent variables—X_n, X_m—are chosen, the other being held constant.

Successful formulation and use of polynomial empirical models requires continuous variation of responses that are rather smooth (with or without transformation) within the local range of interest. Some previous knowledge of the process can provide data regarding any discontinuity of responses. The previous experience also allows proper selection of the process variables such that an optimum (or specified) response is likely to be found. An excellent review of the technique and numerous application examples can be found in textbooks by Box and Draper[1] and Box *et al.*[2] Some aspects of response surface methodology also were used in plasma spraying research.[3]

Response surface methodology can be compared with the Taguchi techniques.[4,5] Both approaches are within the domain of design of experiments for assessment of parametric effects on a particular process. One of the goals of the Taguchi design is minimization of the number of experimental runs, while maintaining the ability for assessment of main factor effects on the response. In this respect, the Taguchi approach is a modified fractional factorial design, with parameter interactions aliased with main effects. Accordingly, the method is excellent for screening a large number of parameters and for isolation of the most important ones.

Another advantage of the Taguchi approach is the ability to analyze signal-to-noise ratios and to minimize the impact of undesirable factors on the process outcome. Taguchi methodology effectively allows improvement of product quality, or makes it possible to produce a "robust" product, relatively unresponsive to uncontrolled variations in process parameters. In this context, response surface methodology is complementary to the Taguchi methodology. However, graphic representation of the response surfaces provides invaluable insight into process variation as a function of the most important parameters, chosen, perhaps, by the Taguchi approach or partial factorial design from a large number of possible variables.

This article illustrates how a number of overlapping response surfaces Y_1, $Y_2 \ldots Y_5$ can be used to select the operating conditions necessary to achieve desired specifications and for optimization of the plasma spraying process. A well-known

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Table 1 Experimental Conditions According to 2^3 Factorial Design
(Runs 1 to 8), plus the center point repeated twice

Run No.	X_1: H_2 in Ar/H_2(a), %	X_2: distance (D)(b), in.	X_3: powder feed (F)(c), g/min
1	4	2	30
2	0	5	30
3	0	2	60
4	0	2	30
5	4	5	30
6	4	2	60
7	0	5	60
8	4	5	60
9	2	3.5	45
10	2	3.5	45

(a) ±3%. (b) ±0.1 in. (c) ±3 g/min.

system—WC-12%Co—deposited on mild steel was chosen to focus on the approach and methodology, and to avoid unexpected deposition problems. It should be emphasized that the tests and conclusions refer specifically to the torch and material used in the present study. However, the illustrated approach and methodology of the response surfaces is universal.

The following five coating properties (*i.e.*, the dependent variables Y_i, indicative of the coating quality) were empirically modeled by the response surfaces: Y_1 = roughness, R (μm); Y_2 = erosion rate, E (mg/sec); Y_3 = Rockwell A hardness, H (RA unit); Y_4 = density, d (g/cm^3); and Y_5 = content of decarburized carbide W_2C, W (arbitrary unit), measured in arbitrary units, as a peak height on the X-ray diffraction diagram. The five responses were modeled in terms of the variation in percentage of hydrogen in an Ar/H_2 primary gas mixture, X_1 = %H_2 (%); torch/substrate distance, $X_2 = D$ (inch) and powder feed rate, $X_3 = F$ (g/min). The choice of these three independent variables was a compromise between (1) limited experimental capability and choice of minimum experimentation to achieve objectives, (2) a large number of variables controlling plasma spraying, and (3) identification of the variables that are most likely to be manipulated in practical plasma spraying operation. All efforts were made to keep other possible experimental variables constant during the deposition of coatings.

2. Design of Experiments

The three independent variables (X_1, X_2, and X_3) were initially set at two levels, according to 2^3 full factorial design.[1] The experimental schedule is presented in Table 1. Eight experiments (=2^3) were necessary to explore the variation of all the independent variables at all levels. The main effects and interactions of X_1, X_2, and X_3 on any chosen response Y_j can be evaluated using standard factorial design procedures. An additional central point was chosen at $0.5X_1$, $0.5X_2$, and $0.5X_3$, and the experiments at the central point were repeated twice to evaluate the possible nonlinear effects (*e.g.*, quadratic or cubic) and to determine repeatability of the deposition process.

It was believed at the outset of experimentation that the three independent variables and/or their combinations would have an effect on the five responses (this is the principal benefit of prior knowledge, *e.g.*, choosing a known system as an example). The choice of responses was additionally dictated by imposing the desired coating specification—roughness below 5 μm, erosion rate (under the erosion conditions specified below) below 10 mg/sec, hardness above 40 RA, density above 9 g/cm^3, and less than 7% of WC reacted to W_2C during spraying. This last condition corresponded to the height of the strongest W_2C peak on an X-ray diffraction diagram of less than five arbitrary units (all diffraction data were collected on the same machine and same current and voltage settings). The presence of the carbon-deficient carbide W_2C was considered an indication of partial oxidation of the feed powder during plasma spraying.

3. Experimental Procedure

The test samples (2.5 by 7.5 by 0.3 cm) were cut from the same billet of 3-mm-thick plain steel, then degreased followed by grit-blasting using 100- to 180-grit alumina. The ten runs of plasma spraying were performed using a Metco MB Plasma Spraying System operated by an experienced technician, at a power level of 24 ± 4 kW (I = 500 A and V between 40 and 55 V, depending on the H_2 percentage mixed with the primary argon gas). The coating thickness was maintained at 375 ± 75 μm, and the same batch of powder was used for all coatings. All deposition runs were accomplished within 4 hr of the same day.

The test conditions for coating properties were fixed for all the samples, and average values of the responses were used for further analysis. The surface roughness tests were performed on a Taylor-Robson Profilometer. The erosion rate test was performed using 100- to 180-grit alumina, directed at a 45° angle to the coating surface, 1.5 cm from the coating and at a blast pressure of 6 atm. The sample was weighed before and after the erosion test. A specific time interval was chosen that assured no exposure of the substrate material during the erosion test. Similar precautions had to be undertaken for the hardness measurements. The initial Vickers diamond pyramid tests resulted in penetration of the substrate surface. The standard (spherical) Rockwell A tip resulted in indentations within the coating body. Densities of detached coatings were determined using the liquid displacement technique. X-ray diffractograms were taken using Cu-K_α radiation. The microstructures of coatings were observed at low (optical microscopy) and high magnifications (scanning electron microscopy).

4. Results and Discussion

The average values and standard deviations of the responses Y_1 to Y_5 from the ten experimental runs are presented in Table 2. The effects of three independent variables (X_1, X_2, and X_3) and all combinations of two variables ($X_i \cdot X_k$; i = 1, 2, 3; k = 1, 2, 3) and combination of all three variables ($X_1 \cdot X_2 \cdot X_3$) on each response were analyzed, and statistical significance tests were performed. The effects were defined as parameters b_{ik} of a best-fit polynomial equation relating response Y_j to combinations of independent variables:

$$Y_j = b_0 + b_1 \cdot X_1 + b_2 \cdot X_2 + b_3 \cdot X_3 + b_{12} \cdot X_1 \cdot X_2 + b_{13} \cdot X_1 \cdot X_3 + b_{23} \cdot X_2 \cdot X_3 + b_{123} \cdot X_1 \cdot X_2 \cdot X_3$$

Table 2 Summary of Coatings Characteristics

Run No.	Y_1: roughness (R)(a), μm	Y_2: erosion (E)(b), mg/sec	Y_3: hardness (H)(c), RA	Y_4: density (d)(d), g/cm^3	Y_5: W_2C (W)(e)
1	6.9	1.3	43	6.8	4.0
2	7.6	95	33	6.8	1.5
3	7.3	15	40	6.0	0.0
4	7.6	19	36	7.1	0.0
5	6.6	1.2	44	10.2	4.5
6	7.0	2.2	55	10.2	7.0
7	7.8	111	35	8.1	2.3
8	5.7	1.0	41	7.8	8.0
9	7.8	1.3	38	8.8	2.0
10	7.0	1.8	38	9.0	4.0

(a) ±0.7 μm. (b) ±0.3 mg/sec. (c) ±4 Rockwell A points. (d) ±0.1 g/cm^3. (e) Arbitrary units, ±0.2.

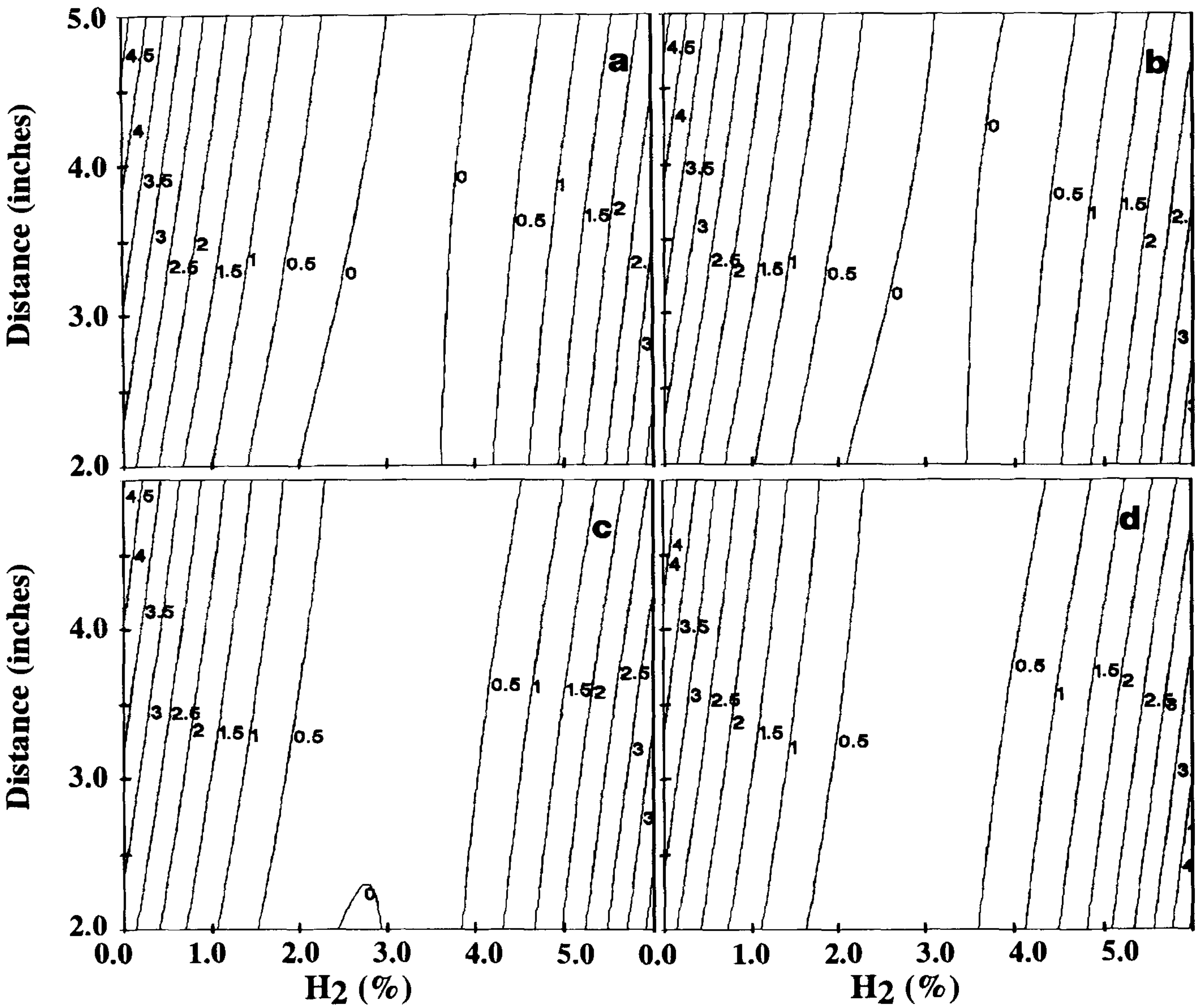

Figure 1 Contours of constant erosion rate (ln E) in mg/sec of plasma-sprayed WC-12%Co coating, as a function of hydrogen content (%) of the primary Ar/H_2 gas, spraying distance (inches), and powder feed (F) of 15 g/min (**a**), 30 g/min (**b**), 45 g/min (**c**), and 90 g/min (**d**).

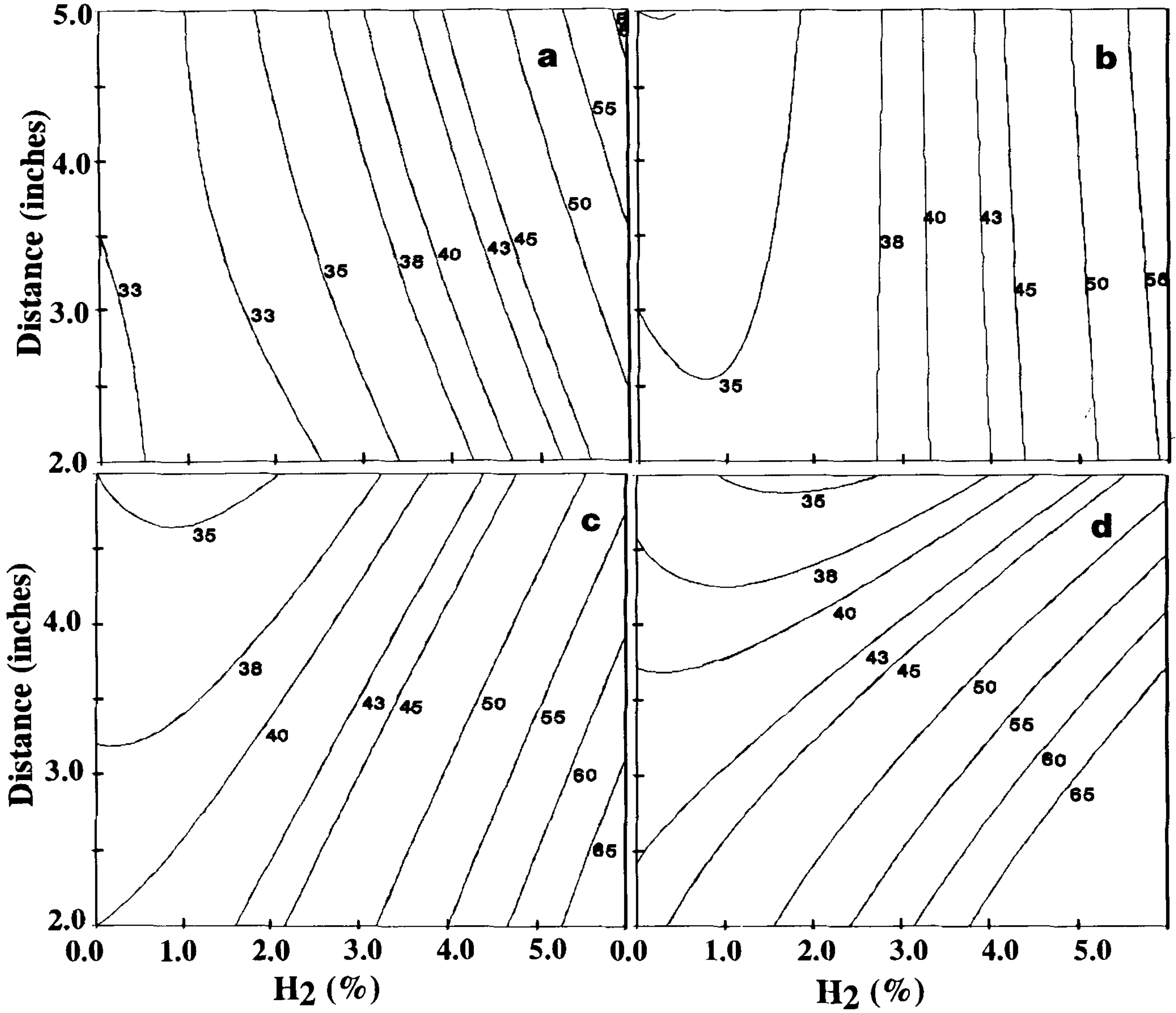

Figure 2 Contours of constant hardness (H) (Rockwell A scale) of plasma-sprayed WC-12%Co coating, as a function of hydrogen content of the primary Ar/H_2 gas, spraying distance (inches), and powder feed (F) of 15 g/min (**a**), 30 g/min (**b**), 45 g/min (**c**), and 90 g/min (**d**).

Only statistically significant parameters were used for determination of the response surface Y_j for each dependent parameter ($j = 1 \ldots 5$). The results of the significance tests are as follows.

Roughness of coatings does not depend on the torch/substrate distance, D, the percentage of H_2, or powder feed rate, F, nor on any combination of these parameters. Equivalently, any variation of roughness measurements results only from experimental error, and the response surface $Y_1 = f(X_1, X_2, X_3)$ is flat. No further modeling of coating roughness was therefore attempted.

Erosion rate depends on $X_1 = \%H_2$, X_2 = spray distance, D, and the combination of the two: $X_1 \cdot X_2$. This is an indication that the erosion rate increases with distance at $H_2 = 0\%$ (examine run No. 3 and No. 4 versus run No. 2 and No. 7 in Tables 1 and 2) and decreases with distance at $H_2 = 4\%$ (examine run No. 1 and No. 6 versus run No. 5 and No. 8 in Tables 1 and 2). A second-degree polynomial $Y_2 = f(X_1, X_2, X_1 \cdot X_2)$ is sufficient to empirically model the erosion rate. The $\ln(Y_2)$ transformation was used to construct the model due to the large variation in the erosion rate. The response surfaces were obtained only in terms of the significant variables X_1, X_2, and $X_1 \cdot X_2$. All the other possible variables and their combinations were dropped from the modeling procedure. Due to the limited number of experiments performed, second-order effects of $X_1 \cdot X_1$ and $X_2 \cdot X_2$ cannot be separately evaluated (these are aliased) and were assumed to be included in the linear effects of X_1 and X_2, respectively.

Hardness depends on all three variables, as well as on the combination of distance and powder feed $X_2 \cdot X_3$ and on the combination of all three variables $X_1 \cdot X_2 \cdot X_3$. Accordingly, coating hardness is a complicated function of the percentage of H_2, distance, and feed and was modeled by a third-degree poly-

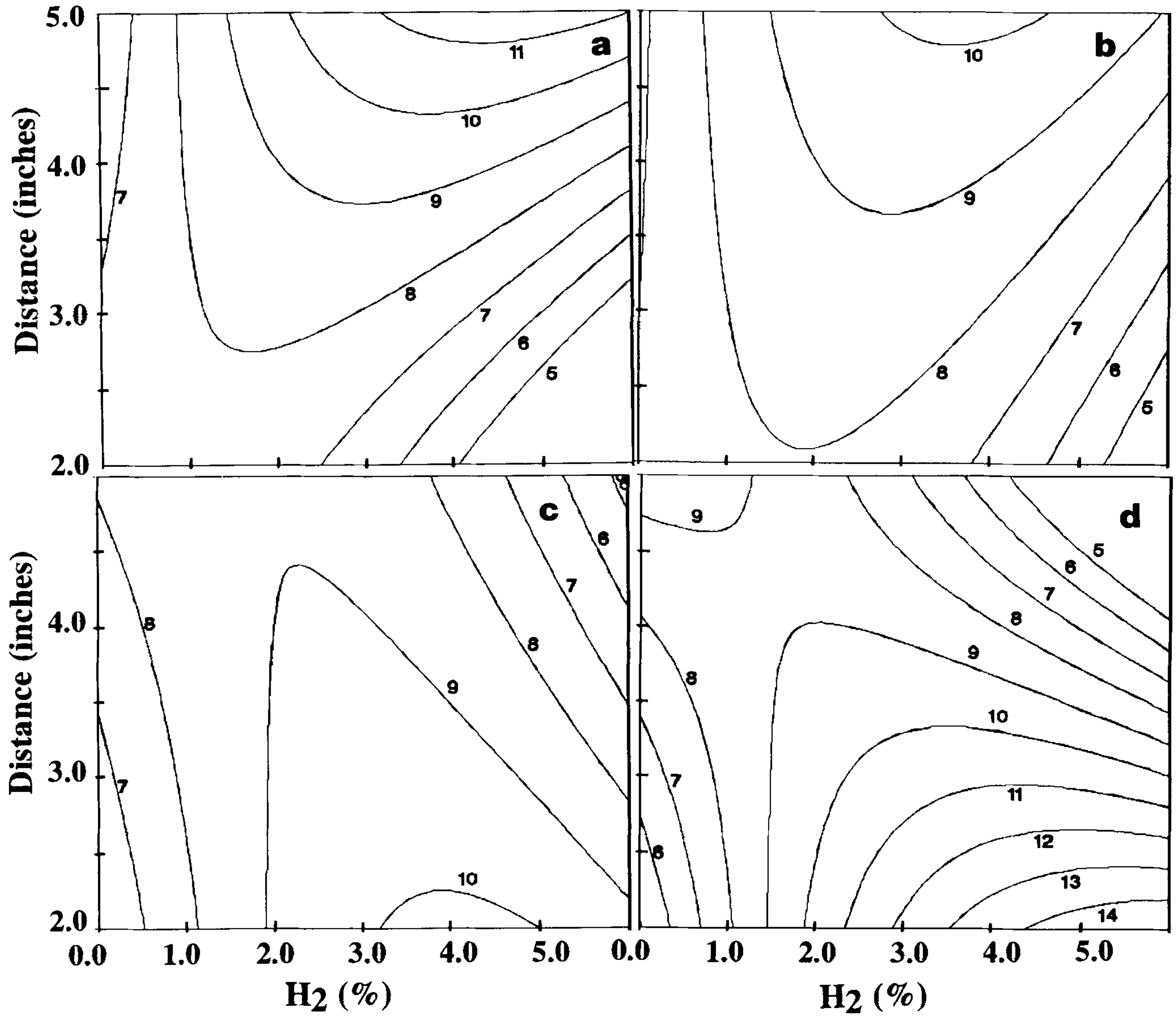

Figure 3 Contours of constant density (d) in g/cm^3 of plasma-sprayed WC-12%Co coating, as a function of hydrogen content (%) of the primary Ar/H_2 gas, spraying distance (inches), and powder feed (F) of 15 g/min (**a**), 30 g/min (**b**), 45 g/min (**c**), and 90 g/min (**d**).

nomial $Y_3 = f(X_1, X_2, X_3, X_2 \cdot X_3, X_1 \cdot X_2 \cdot X_3)$. As above, all square and cubic effects of X_1, X_2, and X_3 are aliased and were not evaluated separately.

The density of the coatings depends on X_1 = %H_2, X_2 = spray distance, D, and the combination of spray distance D and X_3 = feed rate, F, $X_2 \cdot X_3$ and the combination of all three variables $X_1 \cdot X_2 \cdot X_3$. A third-degree polynomial $Y_4 = f(X_1, X_2, X_3, X_2 \cdot X_3, X_1 \cdot X_2 \cdot X_3)$ was necessary to empirically model the dependence of coating density on the hydrogen content, spray distance, and powder feed rate. Aliased effects were excluded from the model.

The amount of carbon-deficient carbide W_2C depends on all three variables X_1, X_2, and X_3 and on the combination of hydrogen content X_1 = percent H_2 and powder feed rate $X_3 = F$. A second-degree polynomial $Y_5 = f(X_1, X_2, X_3, X_1 \cdot X_3)$ was sufficient to empirically model oxidation sensitivity during spraying.

According to the above relationships, second- or third-degree polynomials were least-squares fitted to the respective responses (coating properties). Statistical significance tests were performed on each parameter of the polynomial equation. The significance of models was tested by comparing the sum of squares due to regression (SSR), *i.e.*, total degree of deflection of the response surface $Y = f(X)$ from a flat plane Y = constant, with the sum of squares due to error (SSE), *i.e.*, total difference between measured responses and model-predicted responses. The model was accepted when the determined correlation was better than 95%. To independently evaluate the aliased terms of higher order, an additional eight experiments would have to be performed symmetrically outside the currently chosen range of variables. However, percentage of H_2 cannot be less than zero and $D < 2$ in. might introduce the possibility of overheating of the substrate surface.

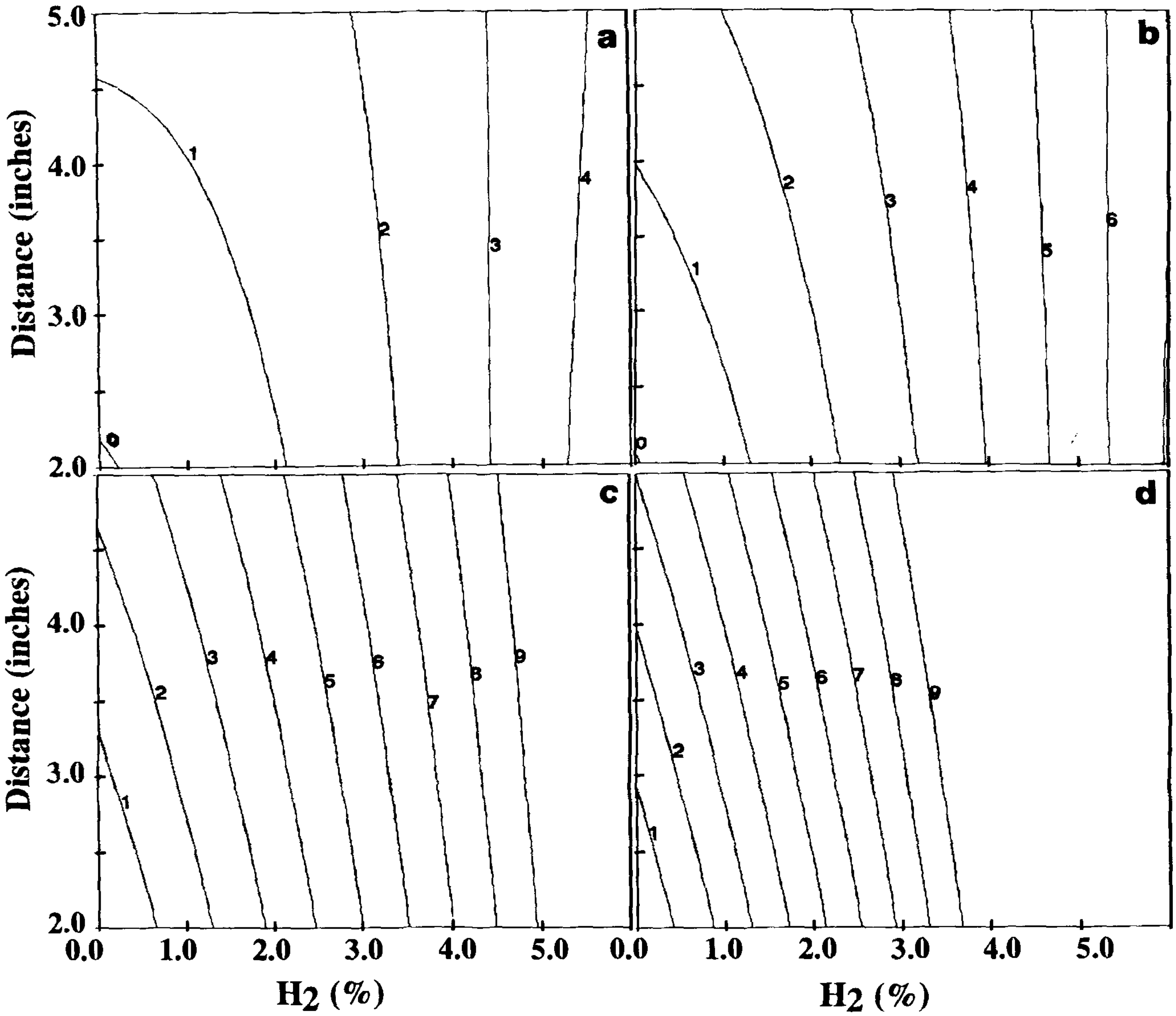

Figure 4 Contours of constant amount of decarburized tungsten carbide W_2C (arbitrary units) in plasma-sprayed WC-12%Co coating, as a function of hydrogen content (%) of the primary Ar/H_2 gas, spraying distance (inches), and powder feed (F) of 15 g/min (**a**), 30 g/min (**b**), 45 g/min (c), and 90 g/min (**d**).

The results of empirical modeling of the coating erosion rate ln(E), hardness, H, density, d, and W_2C content, W, are presented in Fig. 1 to 4, respectively. Each figure is composed of four sections: Section A (predicted for constant powder feed rate, $F = 15$ g/min), Section B (determined for $F = 30$ g/min); Section C (determined for $F = 60$ g/min), and Section D (predicted for $F = 90$ g/min). Each of the sections A to D presents contours of constant response value, within the common axis system: Percent H_2 (%) on the horizontal axis, varied between 0 and 6%, and spray distance, D (inch) on the vertical axis, varied between 2 and 5 in. (10 and 12.5 cm). A degree of prediction (or data extrapolation) is introduced in each figure. Sections A and D present results predicted outside the experimental range. The data for 30 and 60 g/min feed rate values were extrapolated from the measured data. In each section of all figures, the range of percent H_2 = 4 to 6% presents results predicted outside the experimental percent H_2 range of 0 to 4%. The accuracy of model fitting the experiment can be verified by comparison of Fig. 1 to 5 with the data presented in Table 2.

The weak dependence of the erosion rate on powder feed rate is evident from Fig. 1, which presents contours of constant ln (erosion rate) in mg/s. The most durable coatings are produced when sprayed from a short spray distance with a large amount of H_2 in the argon primary gas. This is understandable, because at these conditions, individual splats of higher temperature adhere better to the underlaying solidified layer. An increase in spray distance should be accompanied by an increase in H_2 content to maintain the high temperature of the splats arriving at the coating surface, and thus achieve high resistance to erosion, in that a better formed coating will be produced. Figure 1 predicts deterioration of the erosion resistance as percent H_2 extends beyond 4%, perhaps due to accelerated evaporation of the cobalt binder phase.

In contrast to the erosion rate, rather dramatic changes in hardness versus feed rate are observed in Fig. 2. At low powder feed rates (Fig. 2a and b), the coating hardness is almost independent of the spraying distance, D, and could slightly increase with the spray distance with large amounts of hydrogen in the primary gas, as predicted by Fig. 2(a). As the feed rate increases, hardness starts to decrease with D, at constant percent H_2 (Fig. 2c and d). Thus, to maintain the high hardness of the coating at higher feed rates and larger spraying distances, the amount of hydrogen in the argon should be increased, because this effectively increases the temperature of the sprayed powder. This conclusion is similar, as for erosion resistance and, as one could expect, indicates a proportional relationship between the resistance to erosion and hardness of the plasma-sprayed coatings.

The results of modeling the coating density (Fig. 3) suggest that the torch operated close to the lower limit of power necessary for complete melting of the powder. As the powder feed increases, the spraying distance that results in high density decreases (at constant high percentage of H_2). Some in-flight solidification of the powder is suspected, the amount of solidification being proportional to the flight distance. The highest densities are predicted for 4 to 6% of H_2 at short spraying distances and high powder feed rates.

The amount of powder oxidation, measured as W_2C content and plotted in Fig. 4, is almost linearly proportional to the percentage of H_2, *e.g.*, temperature of the sprayed powder. The isocontours of W_2C content bend slightly toward lower H_2 content with the spraying distance. Time-of-flight between torch and substrate has some influence on the amount of oxidation, because at constant percentage of H_2, more oxidation occurs with increasing spray distance.

Model predictions similar to those presented in Fig. 1 to 5 can serve as a guideline for optimization of the plasma spraying process. To accomplish this task, more verification tests need to be run within areas of the best predicted combination of properties. Based on new experimental results, improved models would be built and the circle of iterations repeated.

The response surfaces presented in Fig. 1 to 4 can be used to define an operating window of the process parameters to achieve a desired coating specification. As an example, the response surfaces were combined in Fig. 5, using conditions that result in a coating erosion rate below 10 mg/sec, hardness above 40 RA, density above 9 g/cm^3, and less than 7% of WC reacted to W_2C (the height of the strongest W_2C peak on X-ray diffraction diagram of less than five arbitrary units). The axis system in Fig. 5 is similar, as in previous figures, but with less extrapolation involved (maximum H_2 = 4.5%). The consecutive contours (*i.e.*, range of distance (D) and percent of H_2 to result in the specified coating properties) are plotted for feed rates (F) of 15, 30, 45, 52, 60, and 75 g/min.

The type of graph shown in Fig. 5 is useful for assessing the stability of the coating properties against the variations in process parameters. A stable (or robust) processing environment allows a large operating window that does not shift substantially even if processing parameters change (intentionally or accidentally). Analysis of Fig. 5 indicates that this is the situation when

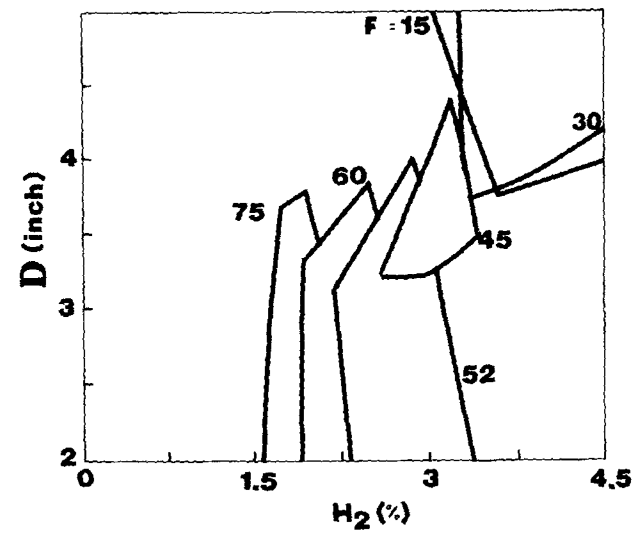

Figure 5 Operating windows of hydrogen content (%) and distance (inches) to achieve coating erosion rate below 10 mg/sec, hardness above 40 RA, density above 9 g/cm^3 and less than 7% of WC reacted to W_2C, for feed rates (F) of 15, 30, 45, 52, 60, and 75 g/min.

spraying proceeds at rather large feed rate ($F = 60 \pm 10$ g/min) with $H_2 = 2 \pm 0.5\%$, at distance (D) of 3 ± 0.5 in., or at a low feed ($F = 20 \pm 10$ g/min), with $H_2 = 4 \pm 0.5\%$, at distance (D) of 4.5 ± 0.5 in. The large feed option is preferable if economy of scale is of concern. However, the powder feed in the range of 40 ± 10 g/min shifts the operating conditions into an unstable region, resulting in coating properties that are sensitive to minor changes in the spraying distance (D) and percentage of H_2.

5. Conclusions

Response surface methodology has been applied successfully to empirical modeling of the plasma spraying process of WC-12%Co. Although this is a well-known coating system, several interesting conclusions follow from the results of modeling. The spraying distance, percentage of H_2 in primary gas, and powder feed rate interact during deposition of coatings. Therefore, investigation of "one factor at a time" to improve the coating process might result in misleading conclusions, and optimal spraying conditions may be missed.

The processing variable that affects the plasma sprayed coating most significantly is the hydrogen content of the primary argon gas. This effect is achieved by changes in the torch power and gas thermal conductivity and enthalpy, leading to higher temperatures and better coating formation. Simultaneously, however, higher H_2 content produces more oxidized coatings. Overlapping of the various response surfaces permits definition of "robust" or optimum conditions for plasma spraying that result in deposit properties that are insensitive to minor variations in processing parameters. For the system under consideration, the optimum processing conditions are feed (F) of 60 ± 10 g/min, hydrogen content of $2 \pm 0.5\%$, and spraying distance (D) of 3 ± 0.5 in.

Acknowledgments

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