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Modelling of Magnetic Proprieties of FeNb Coatings Produced by HVOF Thermal Spraying

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Abstract: In this study, microstructure and magnetic properties of FeNb coatings produced by HVOF thermal spraying were investigated. The as-sprayed coatings were annealed at the temperature of 800°C for 30 min. The results showed that FeNb coatings have nonmagnetic structure. After crystallization by heat treatment, the magnetic properties of the FeNb were weakly improved. Predicted results show that spray distance and fuel flow modified the deposit porosity. A decrease resulted in improvement of coercivity and saturation magnetization

Key words: HVOF, coating, porosity, magnetic properties, modelling, optimization

INTRODUCTION

Magnetic materials underwent a great development in the 20th century (Coey, 2001). Practical progress of magnetism largely depends on relevant advancement in coercivity control resulting from combined control of magneto crystalline an isotropy and microstructure (Coey, 2001).

Amorphous materials can be used as alternative materials for magnetic material applications. These are obtained by a rapid quenching of metal from liquid to solid state with a cooling speed of about 106 K sec^{-1}. They are characterized by long distance order absence of atomic arrangement and consequently they exhibit interesting mechanical, chemical and magnetic properties (Luborsky, 1983).

However, industrial applications related to these amorphous alloys have been restricted because of difficulties related to bulk material production. Thermal spray can resolve this problem by considering rapid solidification of powder particles under high feed rates. In this study, we have used high-velocity oxy-fuel (HVOF) thermal spray technique. This process is adequate for spraying low and intermediate melting temperature materials (e.g., polymers and metals). It permits to obtain high particle velocities needed for amorphization compared to other spray techniques. In this study, FeNb alloy was chosen as feedstock material for its good aptitude to amorphization (Cherigui *et al*., 2003; 2004a, b). Literature is very poor on the use of such material as a feedstock for thermal spraying. It is well known that microstructure, especially grain size, determine the hysteresis loop of a ferromagnetic material. Accordingly, magnetic softening should occur when structural correlation length or grain size becomes smaller than the ferromagnetic exchange length (Alben *et al*., 1978). However, other factors can be associated to the magnetic softening when using thermal spray technology. These are mainly related to an isotropy of the layered structure, porosity level and phase content modification by evaporation.

In order to resolve the posed problem concerning the law connecting the phenomenon to the considered variables, an experimentation process is necessary. Naturally, during experimentation, various values will be given to the variables planned in order to know the influence of these variations on the phenomenon.

In the same time and in order to quantify the role of the porosity of coating on the magnetic properties of FeNb, a model of data processing is considered based on the statistical methods of experiments planning. Such a methodology is an adequate tool for the study of complex processes with parameter interdependencies. In this context, mathematics formulas are used to relate HVOF process parameters to both porosity and magnetic properties of FeNb coatings. The predicted magnetic properties are then correlated to the porosity level for each material, taking into account the interdependency revealed by the optimized network structure.

MATERIALS AND METHODS

Coating manufacturing: Thermal spraying of $Fe_{50}Nb_{50}$ (+0-44) powders was carried out using a commercial Sulzer Metco CDS HVOF spray system on copper substrates. Two substrate shapes were used: Tubes (Ø22×1 mm) and sheets (70×25×1 mm). A gas mixture of oxygen and methane was used to produce the flame. The subsequent combustion of oxygen and methane produced a nominal flame temperature of 2500 K with a hypersonic

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velocity of about 2000 m sec^{-1}. Experiments were carried out by varying two process parameters, namely spray distance X_1 (distance separating the gun tip from the substrate plan) and methane fuel flow rate, X_2. In addition, the cooling system was selected from either water or air system and thus represented the third variable, X_3. The other parameters were kept to a reference condition as shown in Table 1. After spraying, annealing treatment at 800°C was carried out on samples in order to improve their magnetic properties.

Table 1: HVOF spray parameters

Parameters	Values
Spray gun	CDS 89443
Oxygen gas flow rate (SLPM)	420
Methane fuel flow rate, X2 (SLPM)	145, 200
Nitrogen carrier gas flow rate (SLPM)	20
Powder feed rate (g min^{-1})	35
Spray distance, X1 (mm)	200, 300
Substrate type	
-Copper sheet used with air cooling system,	
-Copper tube used with water cooling system	
Cooling system, X3	water, air

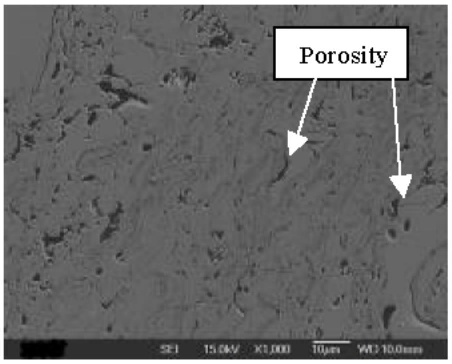

Fig. 1: Morphology of FeNb coatings

Coating characterization: After metallographic preparation, cross-sections of FeNb coatings were analyzed using an optical microscope. The microstructure revealed porosity features presence (Fig. 1).

The percentage of this feature in the microstructure was calculated by image analysis using NIH image free software.

Six images were used to assess mean and standard deviation associated to porosity rate. Magnetic measurements were realized using a hysteresismeter Bull M2000 SIIS, which enabled to draw the hysteresis loop of the considered samples. It permitted also to calculate magnetic properties, namely coercivity Hc and saturation magnetization Ms.

SIMULATION MODEL

The statistical methods of experiments planning used to recognize the correlations between the parameters of a given problem and its responses. The correlations are recognized considering large but simple mathematical operations processed. This technique permits to analyse the processing experimental data; it is effective for the study of the process comprising much independent variable.

The experimentation takes place according to a plan of type $3^1.2^2$ experiences represented on the Table 2 and 3, while varying parameters judged influential. Because of the diversity of parameter units, these are coded according to the relation (1):

Table 2: Varied parameters judged influential

Parameters		- 1	0	+ 1	ΔX_1
Spray distance (mm)	(X1)	200	300.0	400	100.0
Fuel flow (l min^{-1})	(X2)	145	172.5	200	27.5
Cooling type	(X3)	Air	-	Water	-

Table 3: Plan of type $3^1.2^2$ experiences taken

	Input parameters								Output parameters		
	Spray distance	Fuel flow	Cooling type	Interactions					Porosity (%)	Coercivity (Oe)	Saturation magnetization (Am^2 kg^{-1})
Exp. No.	X_1	X_2	X_3	$X_1 X_2$	$X_1 X_3$	$X_2 X_3$	$X_1 X_2 X_3$	$X_1^{\bullet}$	P	Hc	Ms
1	-	-	-	+	+	+	-	1/3	4.2	137	76
2	+	-	-	-	-	+	+	1/3	4.5	136	77
3	-	+	-	-	+	-	+	1/3	4.7	137	76
4	+	+	-	+	-	-	-	1/3	4.1	137	77
5	-	-	+	+	-	-	+	1/3	4.0	137	76
6	+	-	+	-	+	-	-	1/3	4.4	138	76
7	-	+	+	-	-	+	-	1/3	4.0	137	75
8	+	+	+	+	+	+	+	1/3	4.3	138	76
9	0	-	-	0	0	+	0	-2/3	4.2	136	77
10	0	-	+	0	0	-	0	-2/3	4.1	138	75
11	0	+	-	0	0	-	0	-2/3	5.0	136	76
12	0	+	+	0	0	+	0	-2/3	4.4	138	76

(-) inferior value, (0) average value, (+) superior value

$$X_i = (x_i - x_{i0})/\Delta x_i \quad (1)$$

X_i : Input variables
x_i : Real values of input variables
x_{i0} : Basic values of input variables
Δx_i : Interval of variation

RESULTS AND DISCUSSION

Porosity: The confidence interval of the coefficients (Scheffler, 1986):

$|\Delta\beta_i| = S(\beta_i).t_{\alpha, fy} = 0.085$ for $\alpha = 0.05$ and $f_1 = N(m-1) = 24$,

with: $t\,(0.05, 24) = 1.711$ and $S(\beta_i) = 0.05$

$\Delta\beta_i$: Significant value of regression coefficients
β_i : Regression coefficients
$S\{\beta_i\}$: Dispersion of regression coefficients
$t_{\alpha, fy}$: Student test
α : Confidence degree ($\alpha = 0.05$);
f_1 : Freedom degrees number

Considering solely the meaningful regression coefficients, the model will have the shape:

$$P\,(X_i, \beta_i) = 4.37+0.16X_1+0.09X_3+0.09X_1X_3 +0.21X_2X_3-0.09X_1X_2X_3 - 0.187X_1^* \quad (2)$$

After transformation ($X_1^* = X_1^2 - 2/3$), the model becomes:

$$P\,(X_i, \beta_i) = 4.49+0.16X_1+0.09X_3+0.09X_1X_3 +0.21X_2X_3-0.09X_1X_2X_3 -0.187X_1^2 \quad (3)$$

The tentative value of the Fischer criteria (Nalimov *et al.*, 1965) is $F_{exp.} = 2.44$ ($F_{th.} = 2.62$); the model is therefore inadequate.

P : Porosity
$F_{exp.}$: Experimental value of the Fischer test
F_{th} : Theoretical value of the Fischer test

If the spray distance are kept constant for average values ($X_1 = 300$ mm), the model becomes:

$$P\,(X_i, \beta_i) = 4.49 + 0.09X_3 + 0.21X_2X_3 \quad (4)$$

The effect of the fuel flow and the cooling type on the porosity is represented in Fig. 2a.

For the variation interval of fuel flow [157.65, 145] l min^{-1}, the porosity increases nonlinearly in using the air cooling system and decreases in using the water cooling. For a fuel flow variation from 161.5 to 200, the porosity rate increases nonlinearly for the air cooling mode and decreases for the second.

This evolution is due to the reduction of the flame temperature with increasing fuel flow rate (Marple *et al.*, 2001). This reduction favours the presence of the unmolten particles in the coating. In addition, for those particles that do melt their viscosity is increased such that they are unable to impact and adhere to the substrate. Generally, for low fuel flow rates, the particle velocity and temperature are associated with low spray efficiency. For high fuel flow rates, increase of particle velocity and evaporation could be related to the lowering of magnetic property values. These effects are associated with a high porosity level.

If the fuel flow is kept constant for average values ($X_2 = 172.5$ min^{-1}), the model takes the form:

$$P\,(X_i, \beta_i) = 4.49 + 0.16X_1 + 0.09X_3 + 0.09X_1X_3 - 0.187X_1^2 \quad (5)$$

The effect of spray distance and the cooling type on porosity is represented in Fig. 2b.

For a spray distance variation from 200 to 250 mm, the porosity increases linearly and remains stable for a spray distance varying from 250 to 400 mm in using the water cooling system.

This dependence can be explained by considering particle temperature variation with respect to spray distance. For short spray distances, particle residence time is short in the flame. Consequently, they are less heated when they strike the substrate and thus cannot flatten adequately. This leads to a high porosity level in the coating (Sobolev and Giuilemany, 1994; Zhao and Lugscheider, 2004). In contrast, for large spray distances, particles leave the flame and begin to solidify before they impinge on the substrate. The porosity level increases consequently for the same conditions.

Coercivity: The confidence interval of the coefficients (Scheffler, 1986) is equal to 0.24.

Considering the meaningful regression coefficients solely, the model will have the shape:

$$Hc\,(X_i, \beta_i) = 136.53 + 0.13X_1 + 0.58X_2 + 0.25X_1X_2 + 0.38X_1X_3 + 0.6X_1^2 \quad (6)$$

The tentative value of the Fischer criteria (Nalimov and Tschemova, 1965) is $F_{exp.} = 2.20$ ($F_{th.} = 2.62$); the model (6) is therefore inadequate.

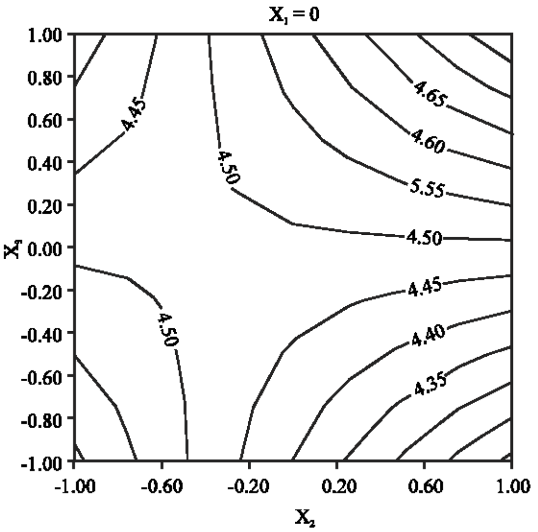

Fig. 2a: Effect of the fuel flow and the cooling type of the porosity

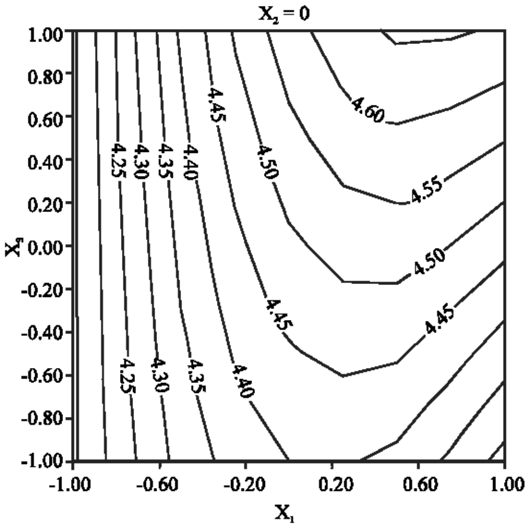

Fig. 2b: Effect of spray distance and cooling type on the porosity

If the spray distance are kept constant for average values ($X_1 = 300$ mm), the model becomes:

$$Hc\,(X_i, \beta_i) = 136.53 + 0.58X_2 + 0.00X_3 \qquad (7)$$

The effect of the fuel flow and the cooling mode on the coercivity is given in Fig. 3a.

From Fig. 3b, we can note that for a spray distance of 300 mm, the fuel flow influences on the coercivity.

When the fuel flow increases, the coercivity shows a clear variation. To explain such a correlation, one has to consider the effect of the coating porosity level as intermediate variable between the process parameters and the magnetic properties.

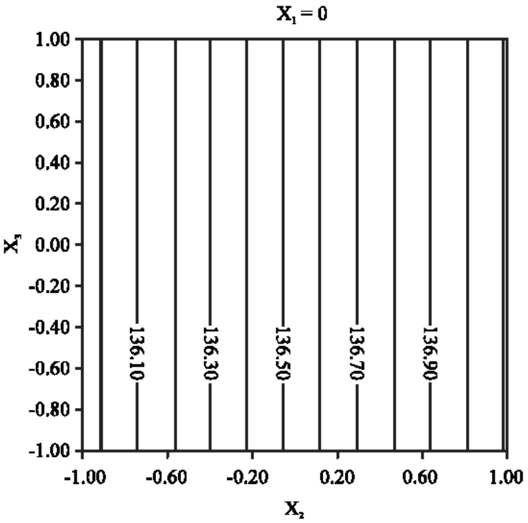

Fig. 3a: Effect of fuel flow and cooling type of the coercivity

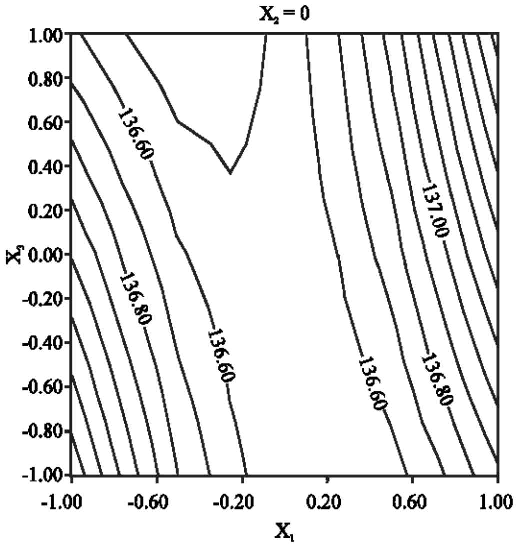

Fig. 3b: Effect of spray distance and cooling type on coercivity

The linear increase of coercivity is explained by the fact that the porosity acts against the continuity of magnetic properties through the coating structure. These are considered as defects anchoring Bloch walls and involving consequently an increase of coercivity (Nacken and Heller, 1961). One can conclude that an improvement in coercivity can be related to low porosity content and this is obtained when spray distance is around 300 mm.

If the fuel flow is kept constant for average values ($X_2 = 172.5\ \text{min}^{-1}$), the model takes the form:

$$Hc\,(X_i, \beta_i) = 136.53 + 0.13X_1 + 0.38X_1X_3 + 0.6X_1^2 \quad (8)$$

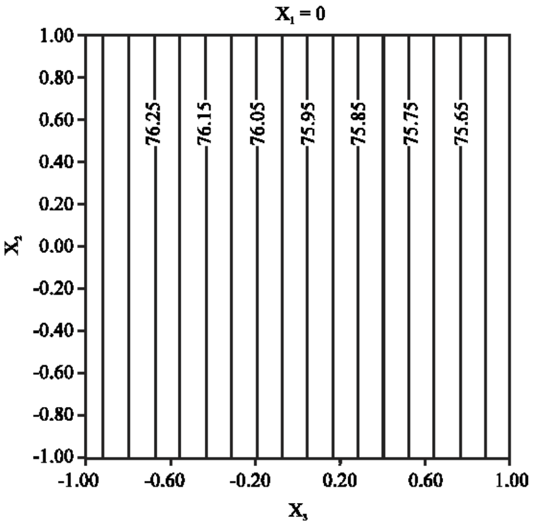

Fig. 4a: Effect of fuel flow and cooling type on the saturation magnetisation

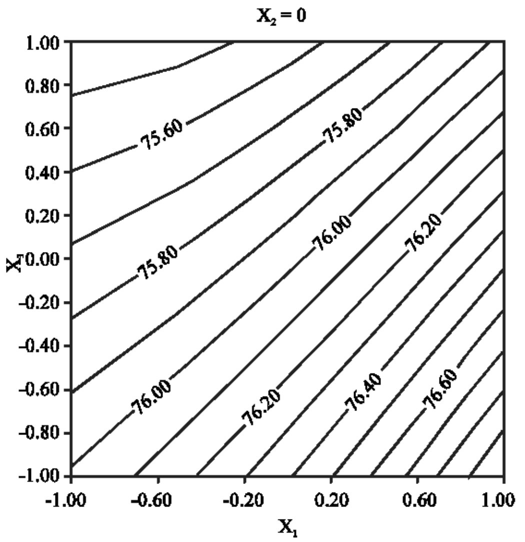

Fig. 4b: Effect of spray distance and cooling type on saturation magnetisation

The effect of spray distance and the cooling type on coercivity is represented in Fig. 3b.

For a fuel flow equal 172.5 l min^{-1}, the coercivity decreases rapidly with the spray distance varying from 200 to 250 mm and slowly from 250 to 280 mm. It increases slowly with the spray distance varying from 310 to 400 mm.

Saturation magnetisation: The confidence interval of the coefficients (Scheffler, 1986) is equal to 0.102.

Considering the meaningful regression coefficients solely, the model will have the shape:

$$Ms\,(X_i,\beta_i) = 75.97 + 0.375X_1 - 0.417X_3 + 0.125X_1X_2 - 0.125X_1X_3 + 0.125\,X_1X_2X_3 + 0.124X_1^2 \qquad (9)$$

The tentative value of the Fischer criteria (Nalimov and Tschernova, 1965) is $F_{exp.} = 2.51$ ($F_{th.} = 2.62$); the model (9) is therefore inadequate.

If the spray distance are kept constant for average values ($X_1 = 300$ mm), the model becomes:

$$Ms\,(X_i,\,\beta_i) = 75.97 + 0.00X_2 - 0.417X_3 \qquad (10)$$

The effect of the fuel flow and the cooling type of the saturation magnetisation is represented in Fig. 4a.

The Fig. 4a shows that for a spray distance of 300 mm, the saturation magnetisation decreases linearly under the effect of the cooling type. In this case the fuel flow does not have any influence.

If the fuel flow is kept constant for average values ($X_2 = 172.5$ min^{-1}), the model takes the form:

$$Ms\,(X_i,\,\beta_i) = 75.97 + 0.375X_1 - 0.417X_3 - 0.125X_1X_3 + 0.124X_1^2 \qquad (11)$$

The effect of spray distance and the cooling type on saturation magnetisation is represented in Fig. 4b.

For a constant value of the fuel flow, the saturation magnetization increases slowly with the spray distance increase. It shows a feeble increase in using the air cooling compared the using of water cooling system.

Generally, in magnetism studies, the decrease of this parameter is related to coercivity increase (Bozorth and William, 1945).

CONCLUSIONS

The research studied the effect of HVOF thermal spraying parameters on the porosity and magnetic properties of coatings using a model of data processing based on the statistical methods of experiments planning:

- The preliminary results showed that FeNb coatings have nonmagnetic structure. After crystallization by heat treatment at a temperature of about 800°C, the magnetic properties of the FeNb were weakly improved.

- Predicted results show that spray distance and fuel flow modified the deposit porosity. A decrease resulted in improvement of coercivity and saturation magnetization.

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