



COMPUTATION AND OPTIMISATION OF ELECTROLESS Ni-Cu-P COATING USING EVOLUTIONARY ALGORITHMS

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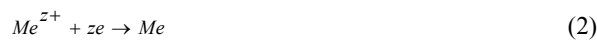
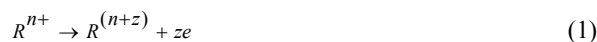
ABSTRACT

Electroless Ni-Cu-P coating was staked on a large concentration of pure Copper substrate. The purpose of present study is to analysis the variation in mass deposition with basis three parameters namely Ni-ion concentration, Cu-ion concentration and reducing agent concentration of the chemical bath. A central composite design of experiments has been considered here as the statistical analysis tool. The mass deposition is treated as the feedback in student's test and it has been found that the concentrations of Ni-ion source and all other interactions significantly influence the mass deposition at 0.05% level of significance. A mathematical model has been developed considering response surface methodology. The optimum concentrations of Ni-ion source, Cu-ion source and reducing agent are obtained using evolutionary algorithms to maximize the mass-deposition per unit area. The coating is again deposited with the optimum concentrations of the parameters and maximum mass-deposition is observed. The XRD of the coatings has revealed that the coating is amorphous in nature.

Keywords: Student's T test, electroless coating, RSM, CCD, F-test, evolutionary algorithms, DE, PSO, ABC

INTRODUCTION

The main component of the electroless bath are the reducing agents, complexing agents, stabilizers operating in a special ion concentration, metal ions, temperature and pH ranges (Reidel W, 1991). The basic chemical reactions (Reidel W, 1991) for the electroless plating are as follows:



This type of metal deposition process is also known as chemical coating process. After the inception of the concept of electroless metal deposition (Reidel W, 1991), the concept has been improved by adding new alloying elements to electroless Ni-P coating bath to develop ternary coatings to meet the need of industry as well as for the purpose of research work. The concentration is focused to the studies of its properties and applications. The main problem of copper is oxidation and wear which can be solved by Ni-Cu-P coating and it can be done keeping the same thermal conductivity and other bulk properties of substrate (Q. Zhao and Y. Liu, 2005). Electroless Ni-Cu-P coating is also applied in VLSI and in thin-film memory discs (Mbouill'eCiss'e *et al.*, 2010). By using gelatin and thiourea as additives in a sulfate solution, Ni-Cu-P can be deposited on 304 stainless steels (N. Parvini-Ahmadi *et al.*, 2004), whereas the corrosion behavior of Ni-Cu-P coating was investigated previously (Guichang Liu *et al.*, 2010). A study of the effect of alloying elements on the amorphous formation of electrolessly deposited Ni-P based coatings (Bangwei and Haowen, 2000) has shown that P is an easily formable

element of the amorphous phase and adding the tin to the binary Ni-P alloys improves the formability of the amorphous phase. In a plating solution, the copper containing electroless Ni-Cu-P alloy coatings mainly depends on Cu-ion concentration, pH and temperature and it has an important effect on the corrosion resistance of the coating (Y. Liu and Q. Zhao, 2004). Combined techniques of reactive DC sputtering of TiN/Ti and electroless deposition of a Ni-Cu-P layer are the most successful techniques of protective hybrid coatings for mild steel preparation. Ni-P coating containing low Phosphorous can transform to the stable phase Ni₃P directly was studied previously, but Ni-Cu-P coating containing low phosphorous but high copper transforms to the metastable phase Ni₅P₂ first, and then to the stable Ni₃P (E. Valova *et al.*, 2010 and Hui-Sheng Yu *et al.*, 2001). Amorphous Ni-Cu-P deposits and the hypereutectic amorphous Ni-P deposits with high phosphorus content first of all transform to the metastable phases Ni₅P₂ and Ni₁₂P₅ and then stable phase Ni₃P (Hui-Sheng Yu *et al.*, 2001). The composition of Ni-Cu-P coating on Aluminium substrate (E. Valova *et al.*, 2005) showed uniformity throughout the thickness, amorphous structure and paramagnetic behavior. The electroless Ni-Cu-P plating improves the brazability of YT15 cemented carbide to steel (Liu Zhu *et al.*, 2012). Cu-ion concentration is the main depending factor for Copper content in electroless Ni-Cu-P-PTFE composite coatings having an important effect on the corrosion resistance of the coatings (Q. Zhao, Y. Liu and E.W. Abel, 2004). When the addition of saccharin is above 4g/L, the behaviour of fracture of the Ni-Cu-P deposit changes from transnodular to internodular. In electroless Ni-B coating process, reducing agent (NaBH₄), metal source (NiCl₂·6H₂O) and temperature significantly affect the deposition (B. Oraon, G. Majumdar and B. Ghosh,



2006). For electroless Ni-B coating at lower bath loading mass deposition becomes significantly high (B. Oraon, G. Majumdar and B. Ghosh, 2007). In electroless Ni-P deposition reducing agent influence both Nickel and Phosphorous content significantly (B. S. Choudhury *et al.*, 2007). Mass deposition per unit area for electroless Ni-Co-P coating depends on individual as well as combined effects of the process parameters which show a significant change in the response variable (T. Banerjee *et al.*, 2012). Ni-Mo-P barrier layers can be deposited on silicon wafers using non-isothermal deposition method without Pd activation (Yu-Hsien Chou *et al.*, 2009). By optimizing few coating parameters viz. annealing temperature, bath temperature, concentration of nickel source and concentration of reducing agent, Friction and wear can be minimized of electroless Ni-B coatings (Suman Kalyan Das *et al.*, 2011 and Prasanta Sahoo, 2009). Nano-particles of MoS₂ which has better friction reduction ability is used in electroless Ni-P coating to minimize friction (Xianguo Hu *et al.* 2009). The P content of the electroless Ni-P deposits increased with the rise of the bath temperature when the rate of electroless Ni-P deposition on 6061 aluminum alloys substrate in an alkaline plating bath and also the coating shows amorphous structure (Wenfeng Qin, 2011). The friction coefficient of Ni-P-PTFE composite coating decreases from 0.33 to 0.12 at 70 N load with an increase in PTFE content from 4.2 to 15.2 wt% (Y. T. Wu *et al.*, 2011). The coating of Ni-P-4.2 wt% PTFE also has good antifriction and wear properties at a load of 30-70 N (Wenfeng Qin, 2011). Sodium dodecyl sulfate surfactant increases corrosion resistance and improves surface morphology of electroless Ni-P coating (Y. T. Wu *et al.*, 2011). Zincate treatment increases the adhesion of the electroless Ni-P coating on various Aluminium alloys (A. Farzaneh *et al.*, 2011). Electroless Ni-P-SiC composite coating on AZ91D magnesium alloy has good anticorrosion property and this coating exhibits an amorphous structure and good adhesion to the substrate (Makoto Hino *et al.*, 2009).

Therefore, it is clear that the properties of electroless Nickel based coatings depends on the concentration of Ni-ion source, reducing agent and other metal ion source, bath temperature etc. Since, no studies have been carried out on the influence of concentration of Ni-ion source, Cu-ion source and reducing agent of Ni-Cu-P coating on mass deposition statistically, therefore, in this present work to understand the role of different process parameters and interactions among those process parameters, the statistical analysis is carried out. With the help of Response Surface Methodology the effects of different parameters are modelled at different levels. The deposited mass per unit area (gm cm⁻²) is considered as the response variable for the statistical analysis and optimization technique and reducing agent concentration, Cu-ion concentration and Ni-ion concentration in the chemical bath are considered as the different variable process parameters. Other parameters such as bath temperature (85°C), concentration of Na₃C₆H₅O₇ · 2H₂O, CH₃COONa · 3H₂O in the chemical bath, activation

temperature (55°C), deposition time (30 minutes), pH (5.64) of chemical bath and bath loading (0.15 cm⁻¹) are kept constant. Also to analyze the microstructure, XRD analysis has been carried out.

SECOND ORDER RESPONSE SURFACE METHOD

From the literature survey it has been observed that, Response Surface Methodology (RSM) is the most efficient tool for improving, developing and optimizing processes by using of statistical and mathematical techniques. In RSM, with controllable negligible error, the independent variables (denoted by X_1, X_2, \dots, X_k) are continuous whereas the response (D) is assumed to be a random variable. This D can be written as a function of X_1, X_2 and X_3 as follows:

$$D = f(X_1, X_2, X_3) + \varepsilon \quad (3)$$

For the required response is $\hat{D} = E(D - \varepsilon)$ then the response surface will be

$$\hat{D} = f(X_1 + X_2 + X_3) \quad (4)$$

To obtain the influence of each process parameters and their interactions, the following lower order that is the first order polynomial equation is considered.

$$y_1 = \beta_0 + \sum_{i=1}^k \beta_i x_i + \varepsilon \quad (5)$$

$$D = \beta_0 + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{123} X_1 X_2 X_3 + \varepsilon \quad (6)$$

The corresponding fitted equation for estimated value of deposited mass can be expressed as follows (B. Oraon *et al.*, 2006, B. Oraon *et al.*, 2007 and B. S. Choudhury *et al.*, 2007):

$$\hat{y}_1 = E(y_1 - \varepsilon) = \hat{\beta}_0 + \sum_{i=1}^k \hat{\beta}_i x_i \quad (7)$$

$$\hat{D} = E(D - \varepsilon) = \hat{\beta}_0 + \hat{\beta}_1 X_1 + \hat{\beta}_2 X_2 + \hat{\beta}_3 X_3 + \hat{\beta}_{12} X_1 X_2 + \hat{\beta}_{13} X_1 X_3 + \hat{\beta}_{23} X_2 X_3 + \hat{\beta}_{123} X_1 X_2 X_3 \quad (8)$$

A second or higher order RSM Model, which is used to approximate the surface around a curvature, can be represented as follows:

$$y = \beta_0 + \sum_{i=1}^k \beta_i x_i + \sum_{i=1}^k \beta_{ii} x_i^2 + \sum_{i=1}^k \sum_{j=1}^k \beta_{ij(i < j)} x_i x_j + \varepsilon \quad (9)$$



The fitted equation is represented by (B. Oraon *et al.*, 2006 and B. S. Choudhury *et al.*, 2007):

$$\hat{y}_2 = E(y_2 - \varepsilon) = \hat{\beta}_0 + \sum_{i=1}^k \hat{\beta}_i x_i + \sum_{i=1}^k \hat{\beta}_{ii} x_i^2 + \sum_{i=1}^k \sum_{j=1}^k \hat{\beta}_{ij(i<j)} x_i x_j \quad (10)$$

$$\hat{D} = E(D - \varepsilon) = \hat{\beta}_0 + \sum_{i=1}^k \hat{\beta}_i X_i + \sum_{i=1}^k \hat{\beta}_{ii} X_i^2 + \sum_{i=1}^k \sum_{j=1}^k \hat{\beta}_{ij(i<j)} X_i X_j + \varepsilon \quad (11)$$

$$\hat{D} = \hat{\beta}_0 + \hat{\beta}_1 X_1 + \hat{\beta}_2 X_2 + \hat{\beta}_3 X_3 + \hat{\beta}_{11} X_1 X_1 + \hat{\beta}_{22} X_2 X_2 + \hat{\beta}_{33} X_3 X_3 + \hat{\beta}_{12} X_1 X_2 + \hat{\beta}_{23} X_2 X_3 + \hat{\beta}_{13} X_1 X_3 \quad (12)$$

EXPERIMENTAL DETAILS

In present study, electroless Ni-Cu-P coating has been deposited on 99.99% pure Copper samples of (20 x 15 x 0.1) mm³ by electroless technique. After cutting from copper foils, required samples are first cleaned by distilled water and then acid pickled in dilute HCL and again rinsed in distilled water. After that the samples are stored to dry. The weight of the substrates has been taken before the coating process on a balance with 0.0001 gm resolution. The copper substrates are then activated by Palladium Chloride solution at 55°C for nucleation of Ni-atom on the surface of the substrate. Then after rinsing them into distilled water, the samples are dipped in the electroless bath which is at 85°C. The electroless bath comprised of the following chemicals as shown in Table-1 and also the conditions of the bath at the time of deposition is mentioned in Table-1. The bath is acidic.

Table-1. Different experimental factors with their ranges (N. Biswas, J. De *et al.*, 2013).

Factors	Range
NiSO ₄ , 7H ₂ O	23.18 – 56.82 gmL ⁻¹
CuSO ₄ , 5H ₂ O	0.86 – 1.54 gmL ⁻¹
NaH ₂ PO ₂ , H ₂ O	25.908 – 46.092 gmL ⁻¹
Na ₃ C ₆ H ₅ O ₇ , 2H ₂ O	55 gmL ⁻¹
CH ₃ COONa, 3H ₂ O	20 gmL ⁻¹
Temperature	85°C
pH	5.64
Time	30 minutes

Minimum 30 minute times are given take for the better deposition of coating and then it again cleaned with distilled water and dried. Then the weight after deposition is taken and compared with the previous weight to calculate the mass-deposition. The mass deposition per unit area is calculated by the following relation (B. Oraon *et al.*, 2006):

Deposition per unit area of the sample,

$$m = \frac{M}{2 \times A} = \frac{M_a - M_b}{2 \times A} = D \times 10^{-4} \text{ (gm cm}^{-2}\text{)} \quad (13)$$

STATISTICAL ANALYSIS

Considering Central Composite Design of experiment, different levels of the independent bath parameters are calculated to determine the significant individual process parameters and interaction, through Student t-test. The actual and coded levels are given in Table-2.

Table-2. Actual and coded values of different process parameters (N. Biswas, J. De *et al.*, 2013)

Actual Values			Coded Values		
NiSO ₄ , 7H ₂ O Z ₁ gmL ⁻¹	CuSO ₄ , 5H ₂ O Z ₂ gmL ⁻¹	NaH ₂ PO ₂ , H ₂ O Z ₃ gmL ⁻¹	X ₁	X ₂	X ₃
30	1.0	30	-1	-1	-1
40	1.2	36	0	0	0
50	1.4	42	1	1	1

The design of experiment is prepared with the help of MINITAB 13.1 software. The experiments are

conducted randomly and then the data are collected after 30 minutes of autocatalytic deposition.

**Table-3.** Data collection for deposited mass per unit area.

Sequence No.	Run No.	Coded Value of NiSO ₄ ·7H ₂ O (gmL ⁻¹)	Coded Value of CuSO ₄ ·5H ₂ O (gmL ⁻¹)	Coded Value of NaH ₂ PO ₂ ·2H ₂ O (gmL ⁻¹)	Average Deposited Mass/unit area, D (gmcm ⁻²)×10 ⁻⁴
1	14	0	0	1.682	48.633333
2	9	-1.682	0	0	16.700000
3	13	0	0	-1.682	53.100000
4	5	-1	-1	1	3.936667
5	1	-1	-1	-1	40.500000
6	11	0	-1.682	0	58.633333
7	2	1	-1	-1	44.966667
8	10	1.682	0	0	49.900000
9	20	0	0	0	59.366667
10	6	1	-1	1	45.966667
11	3	-1	1	-1	38.100000
12	15	0	0	0	58.333333
13	16	0	0	0	60.300000
14	8	1	1	1	47.900000
15	4	1	1	-1	45.466667
16	19	0	0	0	61.500000
17	12	0	1.682	0	54.966667
18	17	0	0	0	59.233333
19	18	0	0	0	62.400000
20	7	-1	1	1	43.133333

DIFFERENTIAL EVOLUTION (DE) ALGORITHM

DE is a population based meta-heuristic algorithm which comprises a population of N candidate solutions in the D -dimensional search space, which at generation t is symbolized by $Pop_t = \{\vec{Z}_1(t), \vec{Z}_2(t), \dots, \vec{Z}_N(t)\}$. (P. Rakshit *et al.*, 2013)

Initialization

The i -th target vector $\vec{Z}_a(t) = \{z_{a,1}(t), z_{a,2}(t), \dots, z_{a,D}(t)\}$ for $a=[1, N]$ at generation $t=0$ is randomly initialized in the range $[\vec{Z}_{\min}, \vec{Z}_{\max}]$ where

$\vec{Z}_{\min} = \{z_{\min-1}, z_{\min-2}, \dots, z_{\min-D}\}$ and $\vec{Z}_{\max} = \{z_{\max-1}, z_{\max-2}, \dots, z_{\max-D}\}$, and thus the b -th component of the a -th member at $t=0$ is selected using

$$z_{a,b}(0) = z_{b-\min} + rand_{a,b}(0,1) \times (z_{b-\max} - z_{b-\min}) \quad (14)$$

For $b = [1, D]$, where $rand_{a,b}$ is a uniformly distributed random number within the range $[0, 1]$. The crossover rate Cr is initialized randomly in $[0, 1]$.

Mutation

Three population members $\vec{Z}_{rand-1}(t)$, $\vec{Z}_{rand-2}(t)$ and $\vec{Z}_{rand-3}(t)$ from the current population Pop_t are selected randomly to create a donor vector $\vec{V}_a(t)$ corresponding to each target vector $\vec{Z}_a(t)$, where

$$\vec{V}_a(t) = \vec{Z}_{rand-1}(t) + U \times (\vec{Z}_{rand-2}(t) - \vec{Z}_{rand-3}(t)) \quad (15)$$

Provided $a \neq rand-1 \neq rand-2 \neq rand-3$ and U is the scaling factor in $[0, 2]$. This is done for $a = [1, N]$.

Crossover

Two types of crossover (recombination) schemes, including binomial and exponential crossover, are found in literature. However, in this paper, binomial crossover is used for generation of a trial vector $\vec{W}_a(t)$ corresponding to each pair of donor vector $\vec{V}_a(t)$ and target vector $\vec{Z}_a(t)$ for the ease of implementation, as given by the following operation.

$$w_{a,b}(t) = \begin{cases} v_{a,b}(t) & \text{if } rand_{a,b} \leq Cr \text{ or } b = b_{rand} \\ z_{a,b}(t) & \text{otherwise} \end{cases} \quad (16)$$



Here $rand_{a,b}$ is a uniformly distributed random number lying in $[0, 1]$ and b_{rand} is an integer index randomly selected from $[1, D]$. This is done for $a = [1, N]$.

Selection

For a given objective $fit(\vec{Z})$ to be maximized, the selection operator, for $a = [1, N]$, is delineated as

$$\begin{aligned} \vec{Z}_a(t+1) &= \vec{W}_a(t) \quad \text{if } fit(\vec{W}_a(t)) \geq fit(\vec{Z}_a(t)) \\ &= \vec{Z}_a(t) \quad \text{if } fit(\vec{W}_a(t)) < fit(\vec{Z}_a(t)) \end{aligned} \quad (17)$$

The steps of mutation to selection are repeated until a termination criterion is reached.

ARTIFICIAL BEE COLONY (ABC) ALGORITHM

A potential solution of an optimization problem is encoded by a food source in ABC (P. Rakshit *et al.*, 2011). In ABC algorithm, the nectar amount of a food source signifies the fitness of the associated trial solution. The number of employed bees and onlooker bees are kept equal to the number of candidate solutions in the population. An algorithm outlining the scheme is discussed below:

Initialization

This phase is identical with initialization phase of DE algorithm. Each food source $\vec{Z}_a(t) = \{z_{a,1}(t), z_{a,2}(t), \dots, z_{a,D}(t)\}$ is assigned a fitness value (nectar amount) $fit(\vec{Z}_a(t))$ for $a = [1, N]$.

Employed Bee Phase: An employed bee produces a modification $\vec{Z}'_a(t)$ on the position (solution) in her memory $\vec{Z}_a(t)$ depending on the local information as stated by (18) and tests $fit(\vec{Z}'_a(t))$, the nectar amount of the new source.

In order to find a solution $\vec{Z}'_a(t) = \{z_{a,1}(t), z_{a,2}(t), \dots, z'_{a,b}(t), \dots, z_{a,D}(t)\}$ in the neighborhood of $\vec{Z}_a(t) = \{z_{a,1}(t), z_{a,2}(t), \dots, z_{a,b}(t), \dots, z_{a,D}(t)\}$, with $b \in [1, D]$ and $c \in [1, N]$, $c \neq a$ are selected on random basis. The value of $z'_{a,b}(t)$ parameter in $\vec{Z}'_a(t)$ solution is computed using the following expression:

$$z'_{a,b}(t) = z_{a,b}(t) + F \times (z_{a,b}(G) - z_{c,b}(G)) \quad (18)$$

Here U is the scale factor in $[-1, 1]$. If $fit(\vec{Z}'_a(t)) > fit(\vec{Z}_a(t))$, the bee memorizes the new position $\vec{Z}'_a(t)$ and forgets the old one i.e. $\vec{Z}_a(t)$. Otherwise she keeps the position of the previous one in her memory. This is done for $a = [1, N]$.

Probability calculation

The probability of each food source $\vec{Z}_a(t)$, for $a = [1, N]$, to be selected by the onlooker bee is computed by

$$prob(a) = \frac{fit(\vec{Z}_a(t))}{\sum_{j=1}^N fit(\vec{Z}_j(t))} \quad (19)$$

Onlooker Bee Phase

A food source $\vec{Z}_a(t)$ is selected by an onlooker bee based on $prob(a)$. Then, following the same principle of foraging behavior of employed bee, onlooker bee also produces $\vec{Z}'_a(t)$ and if $fit(\vec{Z}'_a(t)) > fit(\vec{Z}_a(t))$ bee memorizes $\vec{Z}'_a(t)$ and forgets $\vec{Z}_a(t)$.

Scout Bee Phase

In the ABC algorithm, a food source is abandoned upon unsuccessful trials to improve its fitness through a predefined number of cycles called 'limit'. This abandoned food source is substituted by the scouts by randomly producing a new food source position.

After each evolution, we repeat from employed bee phase until the termination condition is satisfied.

THE PARTICLE SWARM OPTIMISATION (PSO) ALGORITHM

PSO is in principle a multi-agent parallel search technique. Particles are conceptual entities which fly through the multi-dimensional search space (J. Chakraborty and A. Konar, 2008). At any particular instant each particle has a position and a velocity. The position vector of a particle with respect to the origin of the search space represents a trial solution of the search problem.

Initialization

The position $\vec{Z}_a(t) = \{z_{a,1}(t), z_{a,2}(t), \dots, z_{a,D}(t)\}$ and velocity of the a -th particle $\vec{vel}_a(t) = \{vel_{a,1}(t), vel_{a,2}(t), \dots, vel_{a,D}(t)\}$ for $a = [1, N]$ at generation $t=0$ are selected randomly in the range $[\vec{Z}_{min}, \vec{Z}_{max}]$ and $[\vec{vel}_{min}, \vec{vel}_{max}]$ where

$$\vec{Z}_{min} = \{z_{min-1}, z_{min-2}, \dots, z_{min-D}\},$$

$$\vec{Z}_{max} = \{z_{max-1}, z_{max-2}, \dots, z_{max-D}\}$$

$$\vec{vel}_{min} = \{v_{min-1}, v_{min-2}, \dots, v_{min-D}\} \quad \text{and}$$

$$\vec{vel}_{max} = \{v_{max-1}, v_{max-2}, \dots, v_{max-D}\}$$

respectively in the D -dimensional search space. The fitness $fit(\vec{Z}_a(t))$ is evaluated for $a = [1, N]$. The personal best position of the a -th particle is initialized with $\vec{P}_a^{best}(t) \leftarrow \vec{Z}_a(t)$ while the global best position is given as $\vec{G}^{best}(t) \leftarrow \vec{Z}^{best}(t)$. Here



$$\bar{Z}^{best}(t) \leftarrow \arg \left(\max_{\substack{\forall \bar{Z}_a(t), \\ a=[1,N]}} (fit(\bar{Z}_a(t))) \right) \text{ i.e., with highest}$$

fitness. Initialize the inertial weight factor ω .

Velocity update

The velocity of the a -th particle for $a=[1, N]$ is updated as follows:

$$\begin{aligned} \overline{vel}_a(t+1) = & \omega \times \overline{vel}_a(t) + C_1 \times \varphi_1 \times (\bar{P}_a^{best}(t) - \bar{Z}_a(t)) \\ & + C_2 \times \varphi_2 \times (\bar{G}^{best}(t) - \bar{Z}_a(t)) \end{aligned} \quad (20)$$

The first term in the velocity updating formula symbolizes the inertial velocity of the particle. The second term containing $\bar{P}_a^{best}(t)$ represents the best personal experience of each particle and is referred to as “cognitive part”. The last term of the same relation signifies the influence of entire society on the movement of individual particle and hence is interpreted as the “social term” which. Here, C_1 and C_2 are two constant multiplier terms known as “self confidence” and “swarm confidence” respectively. φ_1 and φ_2 are two uniformly distributed random numbers lying within $[0, 1]$ which respectively. A suitable selection of φ_1 and φ_2 govern the degree of influence of $\bar{P}_a^{best}(t)$ and $\bar{G}^{best}(t)$ on the velocity update formula of each particle.

Position Update

The position of the a -th particle for $a=[1, N]$ is updated as follows:

$$\bar{Z}_a(t+1) = \bar{Z}_a(t) + \overline{vel}_a(t+1) \quad (21)$$

Update Personal Best Position

The personal best position of the a -th particle for $a=[1, N]$ is updated as follows:

$$\begin{aligned} \bar{P}_a^{best}(t+1) = & \bar{Z}_a(t+1) \text{ if } fit(\bar{Z}_a(t+1)) \geq fit(\bar{P}_a^{best}(t)) \\ = & \bar{P}_a^{best}(t) \text{ if } fit(\bar{Z}_a(t+1)) < fit(\bar{P}_a^{best}(t)) \end{aligned} \quad (22)$$

Update Global Best Position

The global best position of the swarm is updated as follows:

$$\bar{Z}^{best}(t+1) \leftarrow \arg \left(\max_{\substack{\forall \bar{Z}_a(t+1), \\ a=[1,N]}} (fit(\bar{Z}_a(t+1))) \right) \quad (23)$$

This entire process is iterated from velocity update step until a suitable termination criterion, considering the quality of solution or the upper limit of CPU usage is reached.

RESULTS AND DISCUSSIONS

First Order Polynomial and Student's t Test

Using the Regression analysis in MINITAB release 13.1 statistical software, the coefficients of the factors of the fitted equation are obtained, which can be expressed as

$$\begin{aligned} \hat{D} = & 47.7 + 8.38X_1 + 2.42X_2 - 2.61X_3 - 4.30X_1X_2 \\ & + 5.38X_2X_3 + 4.37X_3X_1 - 5.02X_1X_2X_3 \end{aligned} \quad (24)$$

The estimated ‘t’ values for particular process parameter can be obtained from the following equation:

$$t_{estimated} = \frac{|\text{Coefficient of process parameters}|}{\sigma_\beta} = \frac{|\hat{\beta}|}{\sigma_\beta} \quad (25)$$

$$\text{Now } \sigma_\beta^2 = \sigma_e^2 / n_f \quad (26)$$

$$\sigma_e^2 = \text{Replication variance} = \frac{\sum_{i=1}^{n_c=6} (D_{act} - D_{avg})^2}{(n_c - 1)} = \text{Estimate of error} \quad (27)$$

It is observed that the standard ‘t’ values for 1% and 0.05 % level of significance and 5 degrees of freedom ($v = n_c - 1 = 6 - 1 = 5$) are $t_{0.01;5} = 3.365$ and $t_{0.0005;5} = 6.869$ respectively.

Table-4. Estimated t- values of the process parameters and their interactions.

		Constant	X ₁	X ₂	X ₃	X ₁₂	X ₂₃	X ₁₃	X ₁₂₃
σ_e^2	σ_β	t ₀	t ₁	t ₂	t ₃	t ₁₂	t ₂₃	t ₁₃	t ₁₂₃
2.33052	0.5397	88.382	15.527	4.4839	4.836	7.967	9.6985	8.097	9.3014

From Table-4, it can be concluded that all the main factors and interactions are significant at 1% level of significance and the main factor i.e. concentration of Ni-ion source (X_1) and all the interactions ($X_1 \times X_2$, $X_2 \times X_3$, $X_3 \times X_1$ and $X_1 \times X_2 \times X_3$) are significant at 0.05% level of significance.

Second order response surface and F-test

The coefficients of the fitted equations are obtained through the Response Surface Analysis in MINITAB Release 13.1 statistical software.



$$\begin{aligned} \hat{D} = & 60.43 + 8.38X_1 + 2.42X_2 - 2.61X_3 \\ & -11.08X_1X_1 - 2.77X_2X_2 - 4.87X_3X_3 \\ & -4.30X_1X_2 + 4.37X_1X_3 + 5.38X_2X_3 \end{aligned} \quad (28)$$

The equation is fed to MATLAB 7.0 software to generate the following plots shown in Figure. (1-3).

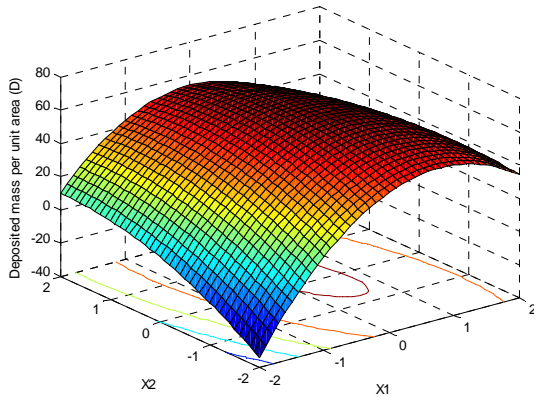


Figure-1. Surface Contour plots for $\hat{D} = f(X_1, X_2)$.

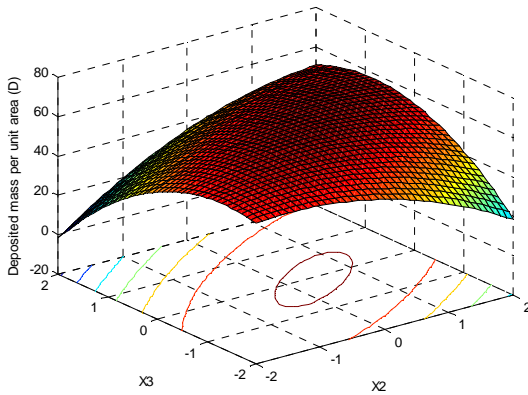


Figure-2. Surface Contour plots for $\hat{D} = f(X_2, X_3)$.

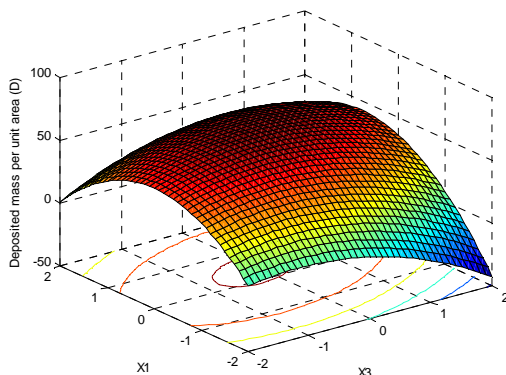


Figure-3. Surface Contour plots for $\hat{D} = f(X_3, X_1)$.

Fisher's Variance Ratio or F-test is used for present study to test of reliability for the predicting response surface equations which is defined as (B. S. Choudhury *et al.*, 2007) (T. Banerjee *et al.*, 2012):

$$F = \sigma_{res}^2 / \sigma_e^2 \quad (29)$$

Where, σ_{res}^2 = Residual variance

$$= \frac{N=20}{\sum_{i=1}^{20} (D_{acti} - D_{esti})^2} (Z - n) \quad (30)$$

It has been observed that the upper degrees of freedom ($v_1 = Z - n$) and lower degrees of freedom ($v_2 = n_c - 1$) are 10 and 5 respectively. The F-value for 0.1% level of significance is $F_{0.001; 10, 5} = 26.92$. The estimated F-value (23.456) is much less than 26.92. Hence it can be concluded that the established predicting equation give an excellent fitting to the observed data. A MATLAB programme is developed to obtain the optimized value of the chemical bath parameters viz. concentrations of Ni-ion source, Cu-ion source and reducing agent using DE, PSO and ABC algorithms. After running the programme the following results are obtained which is shown in Table-5.

Table-5. Results of DE, PSO and ABC algorithms.

Algorithm	NiSO ₄ , 7H ₂ O gmL ⁻¹	CuSO ₄ , 5H ₂ O gmL ⁻¹	NaH ₂ PO ₂ , H ₂ O gmL ⁻¹
DE	56.8200	0.8636	44.7313
PSO	56.8200	0.8636	46.0920
ABC	56.8200	0.8636	46.0920

Electroless Ni-Cu-P coating is again deposited with these optimum concentrations and an average mass-deposition per unit area of $86.5306 \times 10^{-4} \text{ gm cm}^{-2}$ is obtained which is similar to Figures 4-6.

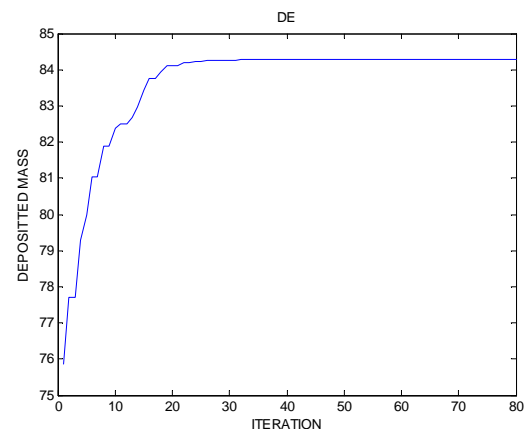


Figure-4. Optimized plot generated by DE algorithm.

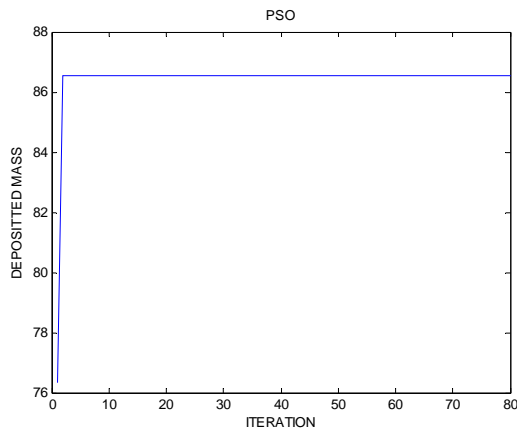


Figure-5. Optimized plot generated by PSO algorithm.

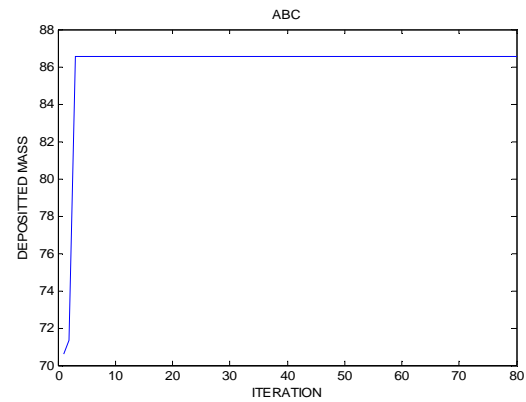


Figure-6. Optimized plot generated by ABC algorithm.

XRD ANALYSIS

Seven as-deposited samples from run no. 9 (-1.682,0,0), 10 (1.682,0,0), 11 (0,-1.682, 0), 12 (0, 1.682, 0), 13 (0, 0, 0), 14 (1, 1, 1) and 20 (-1, 1, 1) are tested through XRD (Rigaku - miniflex, Japan). The samples are mounted on sample holder properly and the samples are tested for XRD analysis for diffraction angle of $0^\circ - 100^\circ$ for 1 hour. Figures 7(a) to (g) shows the results of XRD analysis.

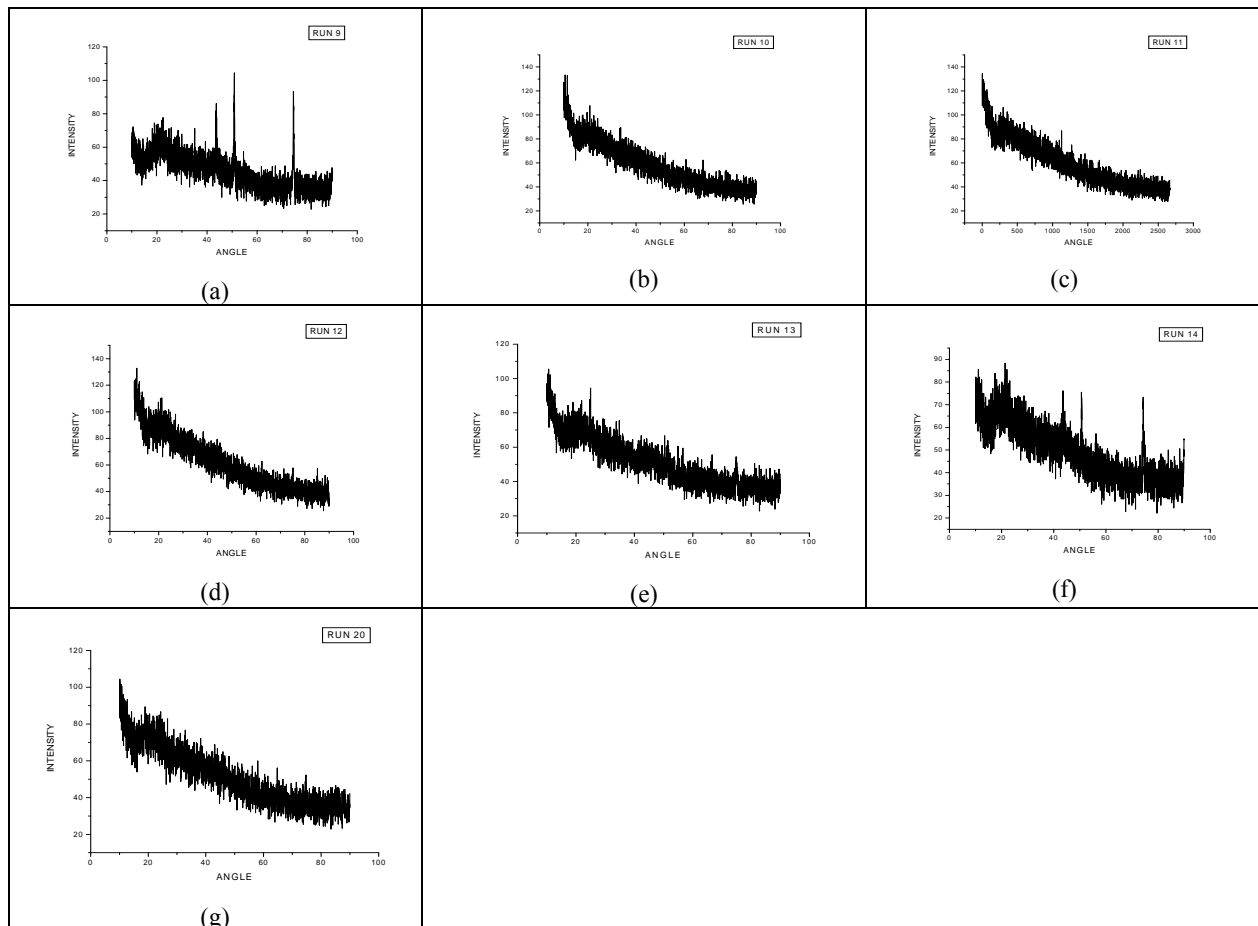


Figure-7. (a) - (g). XRD results of as-deposited coatings.



CONCLUSIONS

The mass deposition per unit area is considered as the response in the student's t test and it can be concluded that the concentration of Ni-ion source and all other interactions significantly influence the mass deposition per unit area at 0.05% level of significance. It has been observed that all main effects and all the interactions of Ni-ion source ($\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$), Cu-ion source ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) and reducing agent ($\text{NaH}_2\text{PO}_2 \cdot \text{H}_2\text{O}$) significantly influence the mass deposition per unit area. The available surface and contour plots also show that the deposited mass per unit area of the substrate is highly influenced by the concentration of nickel ion source in the chemical bath. The deposition also increases up to a certain extent followed by a decrement, when either the concentration of reducing or source of nickel ion increases. The reliability of the predicting response surface equation has been tested by Fisher's F test, which indicates that the established response surface equation is in good agreement with the observed data. After optimizing the process parameters by DE, PSO and ABC algorithms, the coating is again deposited with the optimum concentrations of Ni-ion source ($\text{NiSO}_4 \cdot 7\text{H}_2\text{O}$), Cu-ion source ($\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$) and reducing agent ($\text{NaH}_2\text{PO}_2 \cdot \text{H}_2\text{O}$) and the highest amount of mass-deposition per unit area is observed, therefore, the results of the algorithms converge to the observed data. The diffraction curve related raw data and peak search which is obtained from the computer attached with XRD machine shows that the coating is amorphous.

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Nomenclature

X_i	i-th independent variable
ε	an error component.
β_i	regression co-efficient of the i-th independent variable
β_{ij}	regression coefficient of interaction between i-th and j-th independent variables
β_{ii}	regression coefficient of self-interaction of i-th independent variable
β_{ijk}	regression coefficient of interaction between i-th, j-th and k-th independent variables
$\hat{\beta}_i$	estimated value of β_i
$\hat{\beta}_{ij}$	estimated value of β_{ij}
$\hat{\beta}_{ii}$	estimated value of β_{ii}
$\hat{\beta}_{ijk}$	estimated value of β_{ijk}
E	a mathematical expectation
M	deposited mass per unit area / gcm^{-2}
D	a variable which is $\text{m} \times 10^{-4}$ / gm cm^{-2}
\hat{D}	estimated value of D
Z_i	actual value of the i-th process parameter
$t_{\alpha, \nu}$	value of Student's t- distribution for α level of significance and ν degrees of freedom
σ_β	standard deviation of variability in regression coefficients
l	number of levels
k	number of factors or parameters
n_f	number of factorial points
n_a	number of axial points
n_c	number of axial points
Z	total number of observations = $l^k + 2k + n_c$
n	number of coefficients in the regression equation
$D_{\text{act}i}$	Actual or observed value of deposited mass for the I -th observation
$D_{\text{est}ii}$	Estimated value of deposited mass for the i-th observation
D_{avg}	Average value of deposited mass for the central points
σ_e^2	Replication variance (estimate of error)
σ_{res}^2	Residual variance
F_{α, ν_1, ν_2}	Value of Fisher's F-ratio for α level of significance and ν_1 degrees of freedom ν_2 degrees of freedom
A	Area of one of the two surfaces of the copper sample
M_b	Mass of each copper sample measured before deposition/ gm.
M_a	Mass of each copper sample measured after deposition/ gm.