JOURNAL OF OPTOELECTRONİCS AND ADVANCED MATERİALS Vol. 11, No. 3, March 2009, p. 313 - 319

Empirical modeling of magnetoresistance and electrical resistivity properties of electrodeposited CuCoNi alloys

İSMAIL HAKKI KARAHAN[a*], ÖMER FARUK BAKKALOGLU[b] and H.S. GÜDER[c]
[a]*Department of Physics, Faculty of Arts and Science, Kilis 7 Aralık Universty, 79100, Kilis/Turkey*
[b]*Department of Physics Engeneering, University of Gaziantep, Gaziantep/Turkey*
[c]*Department of Physics Mustafa Kemal University, Hatay/Turkey*

This article presents Genetic Programming (GP) as a new tool for the formulations of magnetoresistance and electrical resistivity properties of electrodeposited CuCoNi alloys. There are no well established formulations for predicting magnetoresistance and electrical resistivity properties of electrodeposited alloys related to film composition. Therefore, the objective of this paper is to develop robust formulations based on the experimental data and to verify the use of GP for generating the formulations for magnetoresistance and electrical resistivity of electrodeposited CuCoNi alloys. To generate databases for the magnetoresistance and electrical resistivity formulations training and testing sets in total of 144 samples were selected at different temperatures and ratios of components. The training and testing sets consisted of randomly selected 123 and 21 for magnetoresistance formulation and 115 and 29 for the electrical resistivity, respectively. The paper shows that the GP based formulation appears to well agree with the experimental data and this is found to be quite reliable.

(Received December 23, 2008; accepted March 19, 2008)

Keywords: CuCoNi, Magnetoresistance, Electrodeposition

1. Introduction

Magnetoresistive materials have recently attracted attention due to their potential technological applications in magnetic data storage and reading. In the presence of magnetic field these materials exhibit large drop in resistivity. This dropping resistivity so-called giant magnetoresistance (GMR) [1,2] effect first observed in Fe/Cr [3] multilayers has made these materials attractive due to their application in magnetoresistive devices.

Different techniques have been used to produce heterogeneous alloys although the structure and therefore properties depend closely on the preparation techniques [4-7]. Electrodeposition, which is a relatively cheap technique, is an alternative method to other complex and sophisticated ones such as evaporation, sputtering, Molecular Beam Epitaxy and it is also suitable for producing multilayer and immiscible metal combinations by control of the electrodeposition variables. Inhomogeneous CuCoNi alloy films are among the systems exhibiting GMR. The GMR and the microstructure in Cu_1-xCox (x = 0.06, 0.13, 0.17, 0.19, 0.21, and 0.26) granular films prepared by electrodeposition were reported [8]. In order to improve GMR of Cu–Co systems, alloying addition such as Cr, Fe, Mn and Ni have been tried [9-12]. Among these elements, only Ni addition gives a prospect with an improved GMR ratio, however the reason for this remains unclear. The phase segregation in the Co–Ni–Cu films is not a pure nucleation decomposition or growth process. It is closely connected with the Ni content. In particular, in the Co–Ni–Cu granular films with low Ni content, the magnetoresistance ratio is larger than in Co–Cu granular films. We previously report an experimental study on the crystal structure, electrical conduction and magnetic properties of $Cu_{100-y-x}Co_yNi_x$ (x=1.56, 2.20, 2.60, 3.06, 4.05, 5.04, 5.5, 6.55, 11.56) alloy films produced by electrodeposition and the effect of Ni addition on the GMR properties [13].

Influence of additional element on magnetoresistance properties is well known in the literature. However, there exist no explicit formulations for estimating the magnetoresistance properties of electrodeposited alloys related to magnetic component like Nickel. For this purpose, empirical formulations were proposed by applying the genetic programming for prediction of magnetoresistance and electrical resistivity of CuCoNi alloys.

2. Experimental details

Electrodeposition of Cu–Co-Ni films was carried out at a constant current density from an aqueous electrolyte of sulfates of Cu, Co and Ni. The electrolytic bath was composed of $CuSO_4.7H_2O$, $CoCl_2.6H_2O$, $NiSO_4.6H_2O$, H_3BO_3, $MgSO_4.7H_2O$, $CoSO_4.7H_2O$, $Na_3C_6H_5O_7.2H_2O$. The substrates for the electrodeposition were aluminum which was subsequently stripped from the films by using 10% NaOH solution. The deposition was performed with a current density of 5 mA/cm^2 at room temperature. A two-electrode composition was used: platinum as anode and aluminum as cathode. The compositions of the films were determined using both an energy dispersive spectrophotometer and an atomic absorption spectrophotometer. The resistivity measurements were determined using the usual four-point probe method in an applied field of ±8.5 kOe using a Varian V- 2900 electromagnet for the temperature depended investigation. The current of 0.1 mA was constant, and directed to the same direction of the magnetic field parallel to the film plane. A helium cryostat (Leybold RW2 Closed Helium Cryostat) was used to control the temperature variation with a sensitivity of ±0.2 K. The dimensions of the samples for the resistivity measurements were 4mm x 4 mm. The magnetic field-MR measurements were carried out in an alternating magnetic field as 0.2, 0.4, 0.6, 0.8, 1.0, and 1.2 at room temperature. The temperature depended MR measurements were carried out in ±8.5 kOe at 20 to 320 K.

3. Genetic programming

Genetic programming was proposed by Koza [14] to automatically extract intelligible relationships in a system and has been used in many applications such as symbolic regression [15, 16] and classification [17, 18]. A schematically overview of genetic programming is given in Fig. 1. Koza [14] explains the flowchart of GP in four main steps:

1. Generate an initial population of random compositions of the functions and terminals of the problem (computer programs)
2. Execute each program in the population and assign it a fitness value according to how well it solves the problem.
3. Create a new population of computer programs.
   - Copy the best existing programs (reproduction)
   - Create new computer programs by mutation
   - Create new computer programs by crossover (sexual reproduction)
   - Select an architecture-altering operation from the program stored so far.
4. The best computer program that appeared in any generation, the best so far solution, is designated as the genetic result of genetic programming.

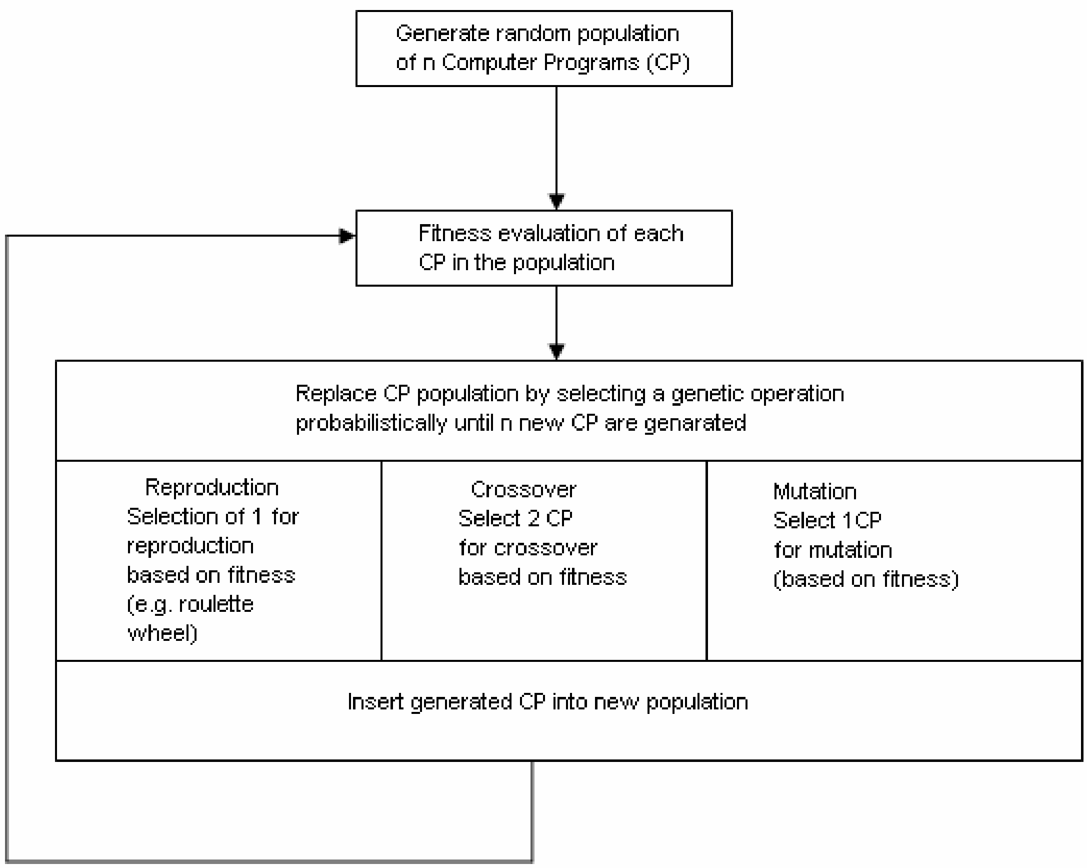


Fig. 1 Schematically overview of GP [19].

The GP creates a population of computer programs with a tree structure. In this study, empirical models are used for prediction of magnetoresistance properties of electrochemically deposited CuCoNi alloys. Randomly generated programs are general and hierarchical, varying in size and structure. GP's main goal is to solve a problem by searching highly fit computer programs in the space of all possible solutions. This aspect is the key for finding near global optimum solutions by keeping many solutions that may potentially to be close to minima (local or global). The creation of initial population is a blind random search of the space defined by the problem. The output of the GP is a program rather than a quantity [20].

3.1 Brief overview of gene expression programming

Gene-Expression Programming (GEP) is a natural development of GP and it was invented by Ferriera [21]. GEP evolves computer programs of different sizes and shapes encoded in linear chromosomes of fixed length. GEP algorithm begins with the random generation of the fixed-length chromosomes of each individual for the initial population. Then the chromosomes are expressed and the fitness of each individual is evaluated based on the quality of the solution it represents [22].

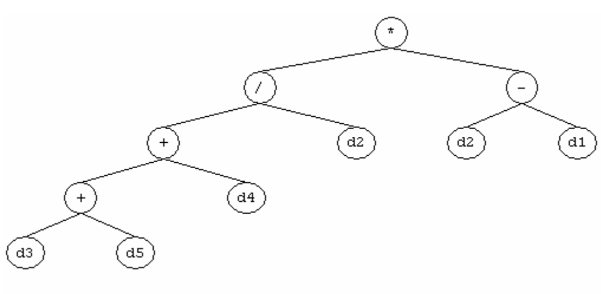


Fig. 2. A typical tree structure for $(((d3+d5)+d4)/d_2)*(d_2-d_1)$

Chromosomes and expression trees (ETs) are the two main parameters of GEP. The process of information decoding (from the chromosomes of the ETs) is called translation which is based on a set of rules. The genetic code is very simple where there exist one-to-one relationship between the symbols of the chromosome and the function or terminal they represent. GEP program utilizes two different languages: the language of genes and the languages of ETs. A noteworthy advantage of this is that it permits the user to infer exactly the phenotype given the sequence of a gene and vice versa; this is called Karva notation [22]. A typical program, representing the expression $(((d3+d5)+d4)/d_2)*(d_2-d_1)$ is shown in Fig. 2.

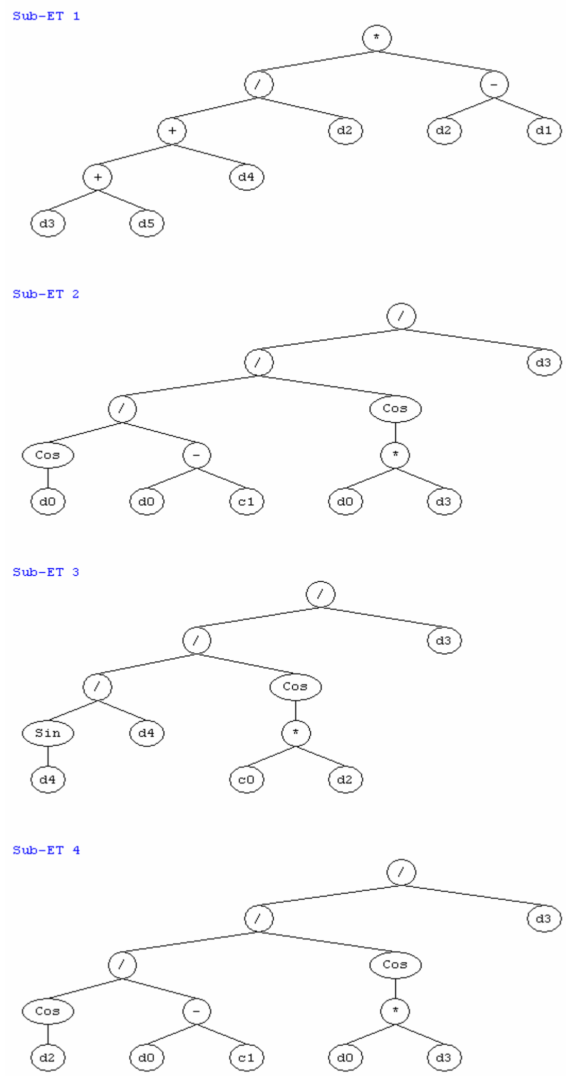


Fig. 3. Tree structure for magnetoresistance of CuCoNi alloys.

4. Application of genetic programming (gep)

The database built in the experimental part was used for the modeling of the magnetoresistance properties of electrodeposited CuCoNi alloys. The major task herein is to define the hidden function connecting the input variables (X1, X2, X3, X4, X5 and X6) and output Y1 and Y2. The expected empirical models may be written in the form of following equation

$$Y_i = f(X1, X2, X3, X4, X5, X6) \qquad (1)$$

The functions obtained by GEP will be used for estimating the relationship between film components and magnetoresistive and electrical resistivity characteristic of CuCoNi alloys. The variables used in the GEP models were presented in Table 1.

Table 1.The variables used in model constructions.

Code	Input variable	Output
X1	Temperature (K)	Magnetoresistance %
X2	Resistivity (ρ_0)	Electrical Resistivity ($\mu\Omega$cm)
X3	Resistivity under magnetic field (ρ_B)	
X4	% Ni content in the film ($Co_{Ni\%}$)	
X5	% Cu content in the film ($Co_{Cu\%}$)	
X6	% Co content in the film ($Co_{Co\%}$)	

In order to construct empirical models and to show the generalization capability of GEP, the database produced in the experimental part is subdivided into two sets, namely training and test, respectively. The empirical formulations were developed based on the former while the latter was employed to test the proposed models so as to measure their generalization capabilities [22]. Of all 144 alloys, the training and testing sets consisted of randomly selected 123 and 21 mixtures, respectively. It must be kept in mind that the proposed empirical equations are valid for the ranges of training set given in Table 2. The parameters used within the proposed empirical models were given in Table 3. Even though there might be various different combinations of GEP parameters, running the GEP algorithm for all of them requires very long computational time. Therefore, the GEP parameters were selected intuitively to investigate the performance of GEP models to predict the magnetoresistance properties of CuCoNi alloys.

Table 2. Ranges of experimental database used in the proposed GEP models.

Code	Parameter	Min	Max
X1	Temperature (K)	20	320
X2	Resistivity (ρ_0)	0.086	1.411
X3	Resistivity under magnetic field (ρ_B)	0.085	1.41
X4	Ni content in film at % ($Co_{Ni\%}$)	1.6	11.5
X5	Cu content in film at % ($Co_{Cu\%}$)	66.6	78
X6	Co content in film at % ($Co_{Co\%}$)	16.5	31.8
Y1	Magnetoresistance at %	-0.07	-2.11
Y2	Electrical Resistivity ρ ($\mu\Omega$cm)	0.086	1.411

Table 3. GEP parameters used for proposed models.

p1	Number of generation	447223
p2	Function set	+,-,*,/,√, Power, e^x, 10^x, x^2, x^3, $\sqrt[3]{x}$, Sin(x), Cos(x)
p3	Chromosomes	30
p4	Head size	8
p5	Number of genes	4
p6	Linking function	Addition
p7	Mutation rate	0.044
p8	Inversion rate	0.1
p9	One-point recombination rate	0.3
p10	Two-point recombination rate	0.3
p11	Gene recombination rate	0.1
p12	Gene transposition rate	0.1

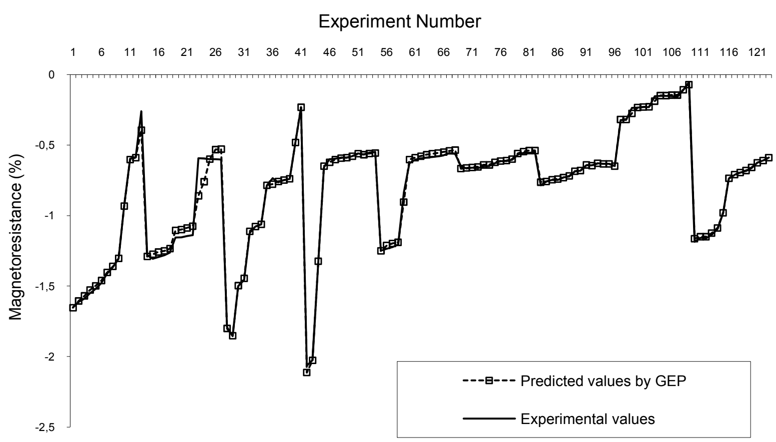
The function generated for the best solutions by GEP algorithm to estimate the magnetoresistance and electrical resistivity predictions of electrodeposited alloys were presented in Equation 2 and 3, respectively

$$MR = ((Co_{Ni\%} + Co_{Cu\%} + Co_{Co\%}) / \rho_B) * (\rho_B - \rho_0)$$
$$+ ((\cos T / (T - 8.171539)) / \cos(T * \rho_B)) / Co_{Ni\%}$$
$$+ ((\sin Co_{Cu\%}) / Co_{Cu\%}) / \cos(-7.872528 * \rho_B)) / Co_{Ni\%}$$
$$+ ((\cos \rho_B / (T - 4.729248)) / \cos(T * Co_{Ni\%})) / Co_{Ni\%}) \qquad (2)$$

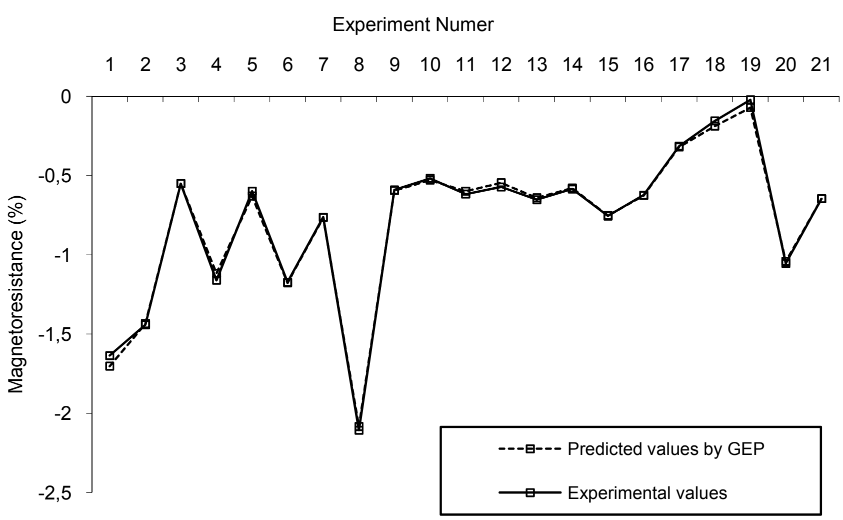
$$\rho = (\cos(Co_{Co\%} + 4.504272 + Co_{Ni\%}))^2$$
$$+ ((((\sqrt{Co_{Ni\%} * T}) * ((Co_{Cu\%} * Co_{Ni\%}) - Co_{Cu\%}))^{1/3})^{1/3})^{1/2}$$
$$+ ((\sin(\sin((-7{,}145294/Co_{Co\%}) + (-6{,}079528)))^2)^{1/3} + (-6.079528))$$
$$+ ((\cos(9.614746 * Co_{Ni\%})) * ((7.264129/Co_{Ni\%}) + Co_{Ni\%}) + Co_{Cu\%})^{1/3} \qquad (3)$$

5. Performance of empirical models

Predicted values achieved through the proposed GEP formulations are compared with the experimental results for the magnetoresistance and electrical resistivity in Figs. 4 and 5, respectively. It was observed in Fig. 4 that the proposed GEP formulation for magnetoresistance of NiCuCo alloys is able to closely follow trend seen in the experimental data within both train and test sets.



(a)

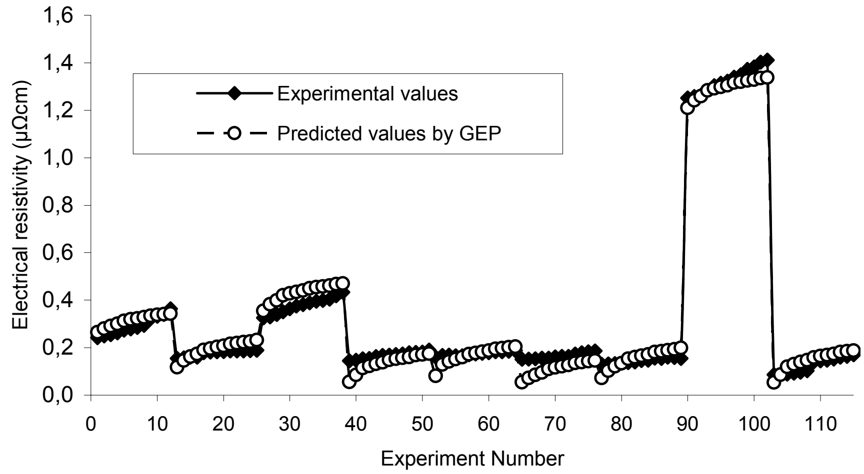


(b)

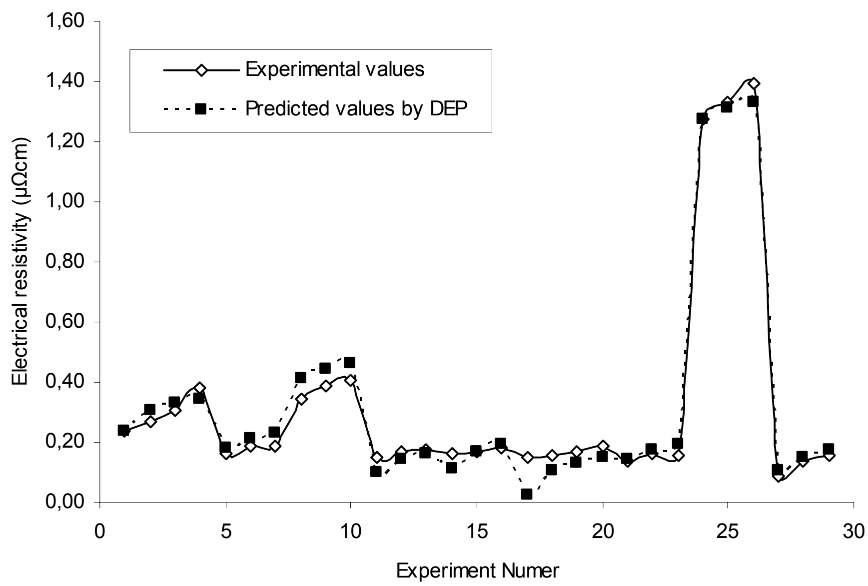
Fig. 4. Evaluation of experimental and predicted magnetoresistance a) Train set b) Test set

It was observed in Fig. 5 that the proposed model for the electrical resistivity provided consistent predictions for both data sets.

The figures fairly showed that there was a clear distinction between the predicted and the actual values when the model was applied to the test set. However, this model well agreed the experimental values in the train set.



(a)



(b)

Fig. 5.Evaluation of experimental and predicted electrical resistivity a) Train set b) Test set.

Table 4. Statistical parameters of GEP formulations.

Properties	Set	MSE	RMSE	MAE	Correlation Coefficient (*R*)
Magnetoresistance	Train	0.00135	0.03679	0.01719	0.99872
	Test	0.00061	0.02482	0.01719	0.99621
Electrical resistivity	Train	0.00145	0.03809	0.03212	0.99459
	Test	0.00178	0.04228	0.03369	0.99278

Statistical parameters of test and training sets of GEP formulations are given in the Table 4 where R corresponds to the coefficient of correlation; MSE is the mean square error, RMSE is the root mean square error; MAE is mean absolute error. As can be seen from Table 4, correlation coefficient of training set of empirical model higher than correlation coefficient of the test set.

6. Conclusions

This paper presents a new and efficient approach for the developing of empirical formulations of magnetoresistance and electrical resistivity properties of electrodeposited CuCoNi ternary alloys. The presented genetic programming approach for modeling the magnetoresistance properties of CuCoNi alloys strongly differs from the conventional methods since it does not use strict mathematical rules and does not derive equations in a rational human way of thinking.

The proposed empirical formulations are based on a comprehensive experimental study.

Because of the high precision of the models developed by the GEP approach, an excessive number of experiments and computations can be avoided, which leads to reduction of the costs of product development. The proposed GEP formulations suggested acceptable agreement with the experimental results. To the knowledge of authors, there exist no explicit formulations for predicting the magnetoresistive properties of electrodeposited CuCoNi alloys in the literature. Therefore, the proposed explicit formulations may be employed in the prediction of the magnetoresistance properties considered in this study.

References

[1] P. M. Levy, Solid State Phys. **47,** 367 (1994).
[2] S. S. P. Parkin, Ann.Rev.Mater. Sci. **25,** 357 (1995).
[3] M. N. Baibitch, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, J. Chazelas, Phys.Rev.Lett. **61**, 2472 (1988).
[4] E. Kneller, J. Appl. Phys. **33**, 1355 (1962).
[5] S. Araki, K. Yasui, Y. Narumiya, J. Phys. Soc. Japan. **60**, 2827 (1991).
[6] Xiao J. Q., Samuel Jiang J., Chien C.L., Phys. Rev. Lett. **68,** 3749 (1992).
[7] M. Alper, K. Attenborough, R. Hart, S. J. Lane, D. S. Lashmore, C. Younes, W. Schwarzacher, Appl. Phys. Lett. **63** 2484 (1994).
[8] O. F. Bakkaloglu, I. H. Karahan, H. Efeoglu, M. Yildirim, U. Çevik, Y. K. Yogurtcu, J. Magn. Magn. Mater **190,** 193 (1998).
[9] M. Rubinstein, V. G. Harris, B. N. Das, N. C. Koon, Phys. Rev. B **50** 17, 12550 (1994).
[10] S. Kondo, T. Yamamoto, K. Yamamoto, T. Morimura, M. Hasaka, J. Appl. Phys. **3**(5), 2453 (1993).
[11] N. Noriyuki, I. J. Kim, H. Takeda, K. Fukamichi, Mater. Sci. Eng. A **181/182**, 88 (1994).
[12] Z. Sun, X. Song, Z. Hu, G. Liang, S. Yang, R. F. Cochrane, J. Alloys and Compaunds. **319**, 276 (2001)
[13] I. H. Karahan, O. F. Bakkaloglu, M. Bedir, Pramana Journal of Physics. **68**, 83 (2007).
[14] Koza Jr. Genetic programming: on the programming of computers by means of natural selection. Cambridge (MA): MIT press; 1992.
[15] Jv Davidson, Da Savic, Ga Walters, Information Sciences **150**(1/2), 95 (2003).
[16] Cs Ong, Huang Jj, Gh. Tzeng, Expert Systems with Applications **29**(1), 41 (2005).
[17] Cd. Stefano, Ad Cioppa, A.Marcell, Pattern Recognition Letters **23**(12), 1439 (2002).
[18] Y. Zhang, S. Bhattacharyya,. Information Science **163**(1/3), 85 (2004).
[19] S. Sette, L. Boullart,. Engineering Applications of Artificial Intelligence **14**(6), 727 (2001).
[20] A. F. Asbour, L. F. Alvarez, V. V. Toropov, Computers and Structure. **81**(5), 331 (2003).
[21] C. Ferreira, Complex Systems; **13**(2), 87 (2001).
[22] A. Cevik, Journal of Constructional Steel Research (2006) article in press.

*Corresponding author:ihkarahan@gmail.com