Journal of Ultrafine Grained and Nanostructured Materials
https://jufgnsm.ut.ac.ir
Vol. 51, No.2, December 2018, pp. 183-192
Print ISSN: 2423-6845 Online ISSN: 2423-6837
DOI: 10.22059/JUFGNSM.2018.02.11

Optimization of micro hardness of nanostructure Cu-Cr-Zr alloys prepared by the mechanical alloying using artificial neural networks and genetic algorithm

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Recieved: 30 October 2017; Accepted: 10 February 2018

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ABSTRACT

Cu–Cr-Zr alloys had wide applications in engineering applications such as electrical and welding industrial especially for their high strength, high electrical as well as acceptable thermal conductivities and melting points. It was possible to prepare the nano-structure of these age hardenable alloys using mechanical alloying method as a cheap and mass production technique to prepare the non-equilibrium materials such as solid solution structures. In this study, artificial neural networks (ANNs) program was developed to establish the relationship between the practical parameters of mechanical alloying, i.e., weight percentages of Cr and Zr as alloying element, milling times, milling speed, sintering time and temperature, on the micro hardness of prepared Cu-Cr-Zr nanostructure alloys. The results of sensitivity analysis showed that the alloying elements and sintering temperature had the highest and lowest effect on the micro hardness of products, respectively. Also, the optimum milling speed and sintering temperature proposed as 255-291 rpm and 530-590°C, respectively. The established models of ANN introduced to genetic algorithm (GA) for determination of the optimal condition. The results were evaluated using the confirmation experiments. Moreover, the optimal condition of nanostructures alloy preparation with the highest micro hardness had been proposed as 310 Hv with the root mean square error (RMSE) of lower than 3.4%.

Keywords: *Artificial neural network; Genetic algorithm; Cu-Cr-Zr nanocomposite; Micro hardness optimization.*

1. Introduction

The characteristics of copper alloy, especially electrical conductivity, converted these alloys as a strategic material in electrical and heat exchanges usages. Relatively, low strength of Cu alloys is one of the most drawbacks for expanding of their application. Solid solution strengthening is one of the most common mechanisms for expanding of the alloy strength. Due to the deterioration effect of large amount of the addition of secondary strengthening element on electrical properties; the amount of alloying element must be optimized to satisfy the electrical properties [1, 2]. Consequently, the maximum strength is restricted using the conventional solid solution strengthening elements. To solve this issue, doping of alloying elements e.g., Zr and Cr with very low solubility, enable us to increase the strength as well as electrical properties in acceptable level. These elements are able to create the significant distortion in very low concentrations [3-5].

Cu–Cr–Zr alloys are generally strengthened using strain hardening as well as precipitation of Cr and Cu-Zr complex phases [6-8]. The possibility of Cu-Zr alloys strengthening using severe plastic deformation is confirmed and optimized in the literatures [2,6,9-11]. Mechanical alloying is another method for the preparations of non-equilibrium phases synthesized includes supersaturated solid solution, metastable crystalline or amorphous alloys, nanostructures and quasi-crystalline phases. Accordingly, the Cu-Cr-Zr alloys prepared through the solid state milling of pure alloying elements, successfully.

Application of optimization strategies on the base of ANN and GA is popular in engineering process, due their unique characteristics for consideration the non-liner relationship between the affecting parameters [12]. ANN technique is able to learn from practical data by determination of weight and estimate the optimum condition, alone. While, GA is a population based evolutionary search as well as optimization process [13, 14]. The usual methodology for combination of this abilities is using of output of ANN as input in GA [13, 15]. This study tried to employ the ANN-GA combination to estimate the optimum condition for maximization of micro hardness of Cu-Cr-Zr alloys. The optimum proposed parameters by GA-ANN is evaluated using the practical experiments.

2. Experimental

The Cu-Cr-Zr alloys were prepared by mechanical alloying of pure elemental powders. The characteristic of the powders are: Cu (Merck, 99.9%, 100 μm), Cr (Merck, 99.8%, 44 μm) and Zr (Merck, 99.8%, 50 μm). The powder mixtures were milled in a planetary ball mill with high chromium stainless steel vial and ball to powder weight ratio (BPR) equal 10:1. To avoid the oxidation of samples, the milling was carried out under the Ar atmosphere. To prevent from temperature raising during the milling operations, 15 min rest was used for every 30 min milling. Crystallinity and phase characterization were determined using XRD. Scanning electron microscopy (SEM) and micro hardness analysis utilized to investigate the morphology and mechanical properties of products. It was necessary to note, by increasing the number of practical parameters in ANN approach, determination of the effect of each parameter was so complicated. To avoid from the complexity of determination the effect of each practical parameters on the micro hardness of products, the effect of other affected parameters (e.g., ligands, stabilizers, volume of cup, atmosphere of furnaces, the size of balls) [16] were ignored and all of them adjusted to a constant value in this study.

Table 1 abbreviates the details of practical parameters as well as the average of micro hardness for every experiment.

Typically, Fig. 1 shows the SEM image and XRD pattern of sample 71. As shown, the prepared Cu-Cr-Zr alloy has flake structure with the average particles size in about 5-80 nm. It was necessary to note that the formation of Cu-Cr-Zr alloy in all sample confirmed by XRD pattern.

2.1 Artificial Neural Network Modeling

ANN methodologies are advanced by brain structure and consist of various parts, i.e., the inputs, outputs, weight vector, activation and transfer functions. These parts situated in three layers, which are input, hidden and output layers. Input layer get input parameters. Hidden layers are responsible for the processing of input layer. The result of hidden layer reported in output layer as output vector, the other constituent of ANN is neuron, i.e., considered as interconnected non-linear memory with less processing elements.

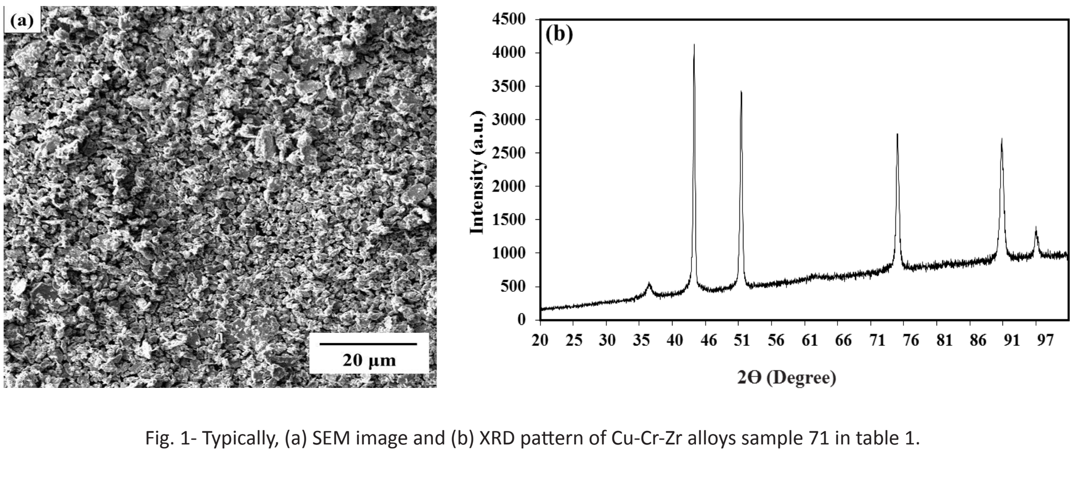

Fig. 1- Typically, (a) SEM image and (b) XRD pattern of Cu-Cr-Zr alloys sample 71 in table 1.

Table 1- Experimental results with inputs and output parameters

No.	Matrix	Reinforcement Cr Wt.%	Reinforcement Zr Wt.%	Bpr	Milling Time (H)	Vial Speed (Rpm)	Sintering Temperature (°C)	Sintering Time (Min)	Micro Hardness (Hv)
1	Cu	0	1	10	20	300	450	30	170
2	Cu	0	1	10	96	300	450	30	200
3	Cu	0	1	10	20	300	600	30	160
4	Cu	0	1	10	40	300	600	30	165
5	Cu	0	1	10	20	300	750	30	100
6	Cu	0	1	10	40	300	750	30	120
7	Cu	0	1	10	96	300	750	30	116
8	Cu	0	1	10	40	300	450	30	175
9	Cu	0	1	10	96	300	600	30	200
10	Cu	0	3	10	20	300	450	30	220
11	Cu	0	3	10	96	300	450	30	280
12	Cu	0	3	10	20	300	600	30	230
13	Cu	0	3	10	40	300	600	30	228
14	Cu	0	3	10	96	300	600	30	228
15	Cu	0	3	10	20	300	750	30	160
16	Cu	0	3	10	40	300	750	30	183
17	Cu	0	3	10	40	300	450	30	232
18	Cu	0	6	10	40	300	450	30	257
19	Cu	0	6	10	20	300	600	30	260
20	Cu	0	6	10	96	300	600	30	190
21	Cu	0	6	10	20	300	750	30	238
22	Cu	0	6	10	40	300	750	30	276
23	Cu	0	6	10	20	300	450	30	260
24	Cu	0	6	10	40	300	600	30	280
25	Cu	1	0	10	12	250	447	30	149
26	Cu	1	0	10	12	250	599	30	106
27	Cu	1	0	10	12	250	753	30	31
28	Cu	1	0	10	48	250	451	30	199
29	Cu	1	0	10	48	250	601	30	200
30	Cu	1	0	10	48	250	751	30	155
31	Cu	1	0	10	96	250	452	30	215
32	Cu	1	0	10	96	250	602	30	222
33	Cu	1	0	10	96	250	751	30	169
34	Cu	3	0	10	12	250	450	30	151
35	Cu	3	0	10	12	250	599	30	127
36	Cu	3	0	10	12	250	750	30	45
37	Cu	3	0	10	48	250	452	30	206
38	Cu	3	0	10	48	250	601	30	243
39	Cu	3	0	10	48	250	750	30	186
40	Cu	3	0	10	96	250	452	30	220
41	Cu	3	0	10	96	250	601	30	242
42	Cu	3	0	10	96	250	752	30	191
43	Cu	6	0	10	12	250	452	30	166
44	Cu	6	0	10	12	250	602	30	134
45	Cu	6	0	10	12	250	751	30	64
46	Cu	6	0	10	48	250	452	30	208
47	Cu	6	0	10	48	250	604	30	189
48	Cu	6	0	10	48	250	751	30	176
49	Cu	6	0	10	96	250	454	30	238
50	Cu	6	0	10	96	250	602	30	189
51	Cu	6	0	10	96	250	756	30	184
52	Cu	0.5	0.5	10	12	300	450	30	111
53	Cu	0.5	0.5	10	48	300	450	30	217
54	Cu	0.5	0.5	10	96	300	450	30	222
55	Cu	0.5	0.5	10	12	300	600	30	100
56	Cu	0.5	0.5	10	48	300	600	30	243
57	Cu	0.5	0.5	10	96	300	600	30	261
58	Cu	0.5	0.5	10	12	300	750	30	97
59	Cu	0.5	0.5	10	48	300	750	30	223
60	Cu	0.5	0.5	10	96	300	750	30	170
61	Cu	1.5	1.5	10	12	300	450	30	134
62	Cu	1.5	1.5	10	48	300	450	30	270
63	Cu	1.5	1.5	10	96	300	450	30	262
64	Cu	1.5	1.5	10	12	300	600	30	114
65	Cu	1.5	1.5	10	48	300	600	30	247

Table 1- Continued

66	Cu	1.5	1.5	10	96	300	600	30	277
67	Cu	1.5	1.5	10	12	300	750	30	111
68	Cu	1.5	1.5	10	48	300	750	30	262
69	Cu	1.5	1.5	10	96	300	750	30	222
70	Cu	3	3	10	12	300	450	30	171
71	Cu	3	3	10	48	300	450	30	309
72	Cu	3	3	10	96	300	450	30	241
73	Cu	3	3	10	12	300	600	30	160
74	Cu	3	3	10	48	300	600	30	247
75	Cu	3	3	10	96	300	600	30	248
76	Cu	3	3	10	12	300	750	30	158
77	Cu	3	3	10	48	300	750	30	220
78	Cu	3	3	10	96	300	750	30	250

Connection of neurons to each other is carried out by weights. These weight help to the network to solve the problem. A connected neuron formula is reported as eq. 1:

$$n = \sum_{i=1}^{p} w_i n + b \qquad \text{(eq. 1)}$$

In which, b is the bias of neurons, p is the number of elements and w_i is the weight of the input vector a_i. Each neuron receives sum of the weight inputs with bias and used from activation function to validate its output signal, as eq. 2:

$$f(n) = a\left(\sum_{i=1}^{p} w_i n + b\right) \qquad \text{(eq. 2)}$$

In which, a is the transfer function of the neuron. Some of the most common transfer functions are log-sigmoid (Logsig: eq. 3); tan-sigmoid (Tansig: eq. 4) and linear transfer function (Purelin: eq. 5).

$$a(n) = \frac{1}{1 + \exp(-n)} \qquad \text{(eq. 3)}$$

$$a(n) = \frac{1}{1 + \exp(-2n)} - 1 \qquad \text{(eq. 4)}$$

$$a(n) = n \qquad \text{(eq. 5)}$$

Fig. 2 schematically shows these transfer function. Feed-forward backpropagaton is one of the most common strategies for training of the network in ANN. This approach enables us to present effective solutions for engineering applications, especially, in material science [17-19].

Detailed explanations of the functions of hidden and output layers are descripted in [17, 20, 21]. As shown in Table 2, by consideration of regression as criteria, “Logsig” and “Purelin” are the best functions for hidden and output layers, respectively.

In this study, feed-forward neural network with back-propagation algorithm employ to simulate the process. In feed-forward strategy, the output of each neuron is merely related to the next layers neurons. This strategy consists of one or more hidden layers and has one output layers. To increase the performance of evolution, all input data normalized in the range 0 - 1. Hidden layers by employment of non-linear transfer function manage the network to learn the linear or non-linear behavior among the input and outputs. Normalizing is done using eq. 6. [21]:

$$X_n = 0.8 \times \frac{X - X_{min}}{X_{max} - X_{min}} + 0.1 \qquad \text{(eq. 6)}$$

In which, X_{max} and X_{min} are the highest and lowest

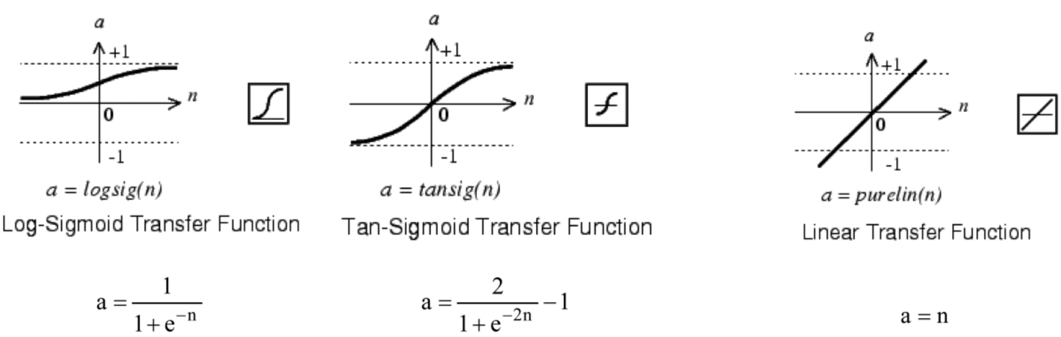

Fig. 2- Schematically representation of transfer functions [16].

values of the independent X variable, respectively. The experimental data divided into two parts randomly. In this work, the back-propagation employed by a network that have an input layer with their neurons for every practical factor (table 1) and an output layer include one neurons (micro hardness); 65 patterns of the experimental results were employed to train the ANN model, and the remaining employed for testing. Linear regression analysis is carried out to compute the correlation coefficient (R^2) between the experimental and predicted values. Fig. 3 illustrates the comparison between the experimental and predicted data. Accordingly, correlation coefficient for training and testing stage are 0.9914 and 0.9834, respectively. The results of neural network confirmed the experimental data by high accuracy.

Various combinations are tested to select the optimum condition. To validate each combination, root mean square error (RMSE) is used as metric and calculated form eq. 7. [21, 22]:

$$\mathrm{RMSE} = \frac{1}{\mathrm{N}}\sum_{1}^{\mathrm{N}}\left(\frac{|\text{Actul value} - \text{Predicted value}|}{\text{Actul value}} \times 100\right) \qquad \text{(eq. 7)}$$

The minimum of RMSE belongs to the network with "Logsig" function for hidden layers, Purelin function for output layer and the number of neurons in hidden layer was (5-30-15-1). Fig. 4 schematically represents the proposed network structure of this study. From table 2, the minimum amount of error is equal to 3.4% and consequently this structure selects for further optimization.

Back propagation is used for learning of network, where comprises from two passes within various layer of network as forward pass and backward pass. In the former, the input vector is used to the network and its effect extended through the network layer by layer. In this stage, the synaptic weights are supposed to be fixed. While, in the later, the parameters of network are fixed and changed to enhance its adaptability to the practical data. Accordingly, the difference of network output and desired value is estimated and the results used in backward pass to update the synaptic weights. The same trend is repeated until the accuracy of prediction would be maximized [23, 24].

Table 2- Amount of regressions for different ANN structure

No.	Activation Function			Neurons In Hidden Layers		Regression (R^2)
	Layer One	Layer Two	Output Layer	Layer One	Layer Two	
1	Tansig	Tansig	Tansig	10	3	0.9464
2	Tansig	Logsig	Purlin	1	14	0.4051
3	Logsig	Logsig	Tansig	5	5	0.5501
4	Logsig	Logsig	Logsig	7	12	0.7959
5	Tansig	Tansig	Purlin	20	4	0.9256
6	Logsig	Tansig	Logsig	3	7	0.8390
7	Logsig	Tansig	Purlin	9	15	0.8929
8	Tansig	Logsig	Tansig	30	27	0.9143
9	Tansig	Tansig	Logsig	16	5	0.7912
*10	Logsig	Logsig	Purlin	30	15	0.9914

*Best Ann Structure

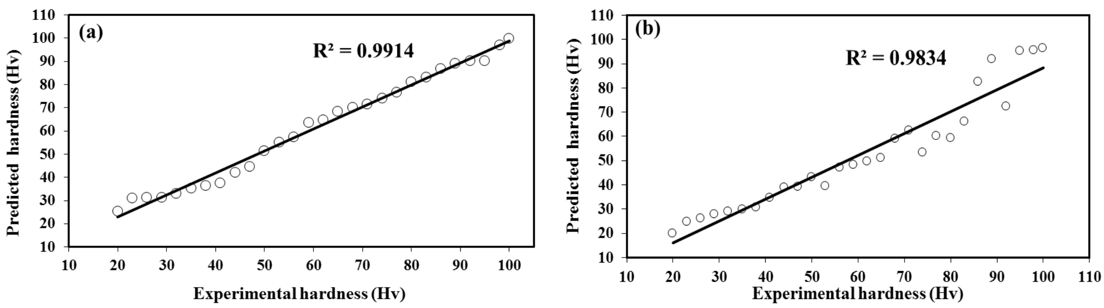

Fig. 3- Regression analysis of predicted and experimental hardness for the best structure: (a) Training data; (b) Testing data.

2.2 Genetic Algorithms (GA)

GA is one of the most common techniques in artificial intelligence on the back of stochastic non-linear optimization strategies without the necessity of extra information, e.g, and gradient. For optimization, GA must be including of selection, crossover and mutation. The first is responsible for the selection of excellent individuals from the current data that they can propagate in the future. The second arrange the parent individuals in pairs, exchanging partial chromosomes among them with a specific probability (i.e., crossover probability), and embedded them to prepare the next generation of new individuals possessing the specification of parent individuals. The thirds is able to select the individuals in the population and manipulate one or a few genes of the selected individuals with the mutation probability to save the generation more diverse and favorable. This trend iterated till repeats several iterations one chromosome has the best fitness and, hence, the chromosome is considered to be as the optimal solution [25-28].

3. Results and Discussion

As shown in Fig. 4, the best ANN structure has 18 neurons in hidden layer with 3.4% error among the 78 structures. Moreover, the predicted and experimental data are close to the each other and consequently confirmed the accuracy of simulation. Sensitivity analysis is employed to determine the most affecting parameters as well as the degree of importance as output parameters. The base of this analysis is the changing of output as a function of changing in each practical parameter. Moreover, the outputs of sensitivity analysis are so helpful for decision about the "robustness" of model parameters that guide the researches to a better decision [29, 30]. RMSE values for different

Table 3- The assumptions made with regards to the simulations

Initial Step Size In Interval Location Step	Default:0.01
Parameter To Avoid Small Reductions In Performance	Default: 0.1
Lower Limit On Change In Step Size	Default: 0.1
Upper Limit On Change In Step Size	Default: 0.5
Maximum Step Length	Default: 100
Minimum Step Length	Default: 1e-6
Maximum Step Size	Default:26
Training Epoch Number	150
Training Error Goal	Default: 0
Initial Step Size	Default: 0.01
Step Size Decrease Rate	Default: 0.9
Step Size Increase Rate	Default: 1.1

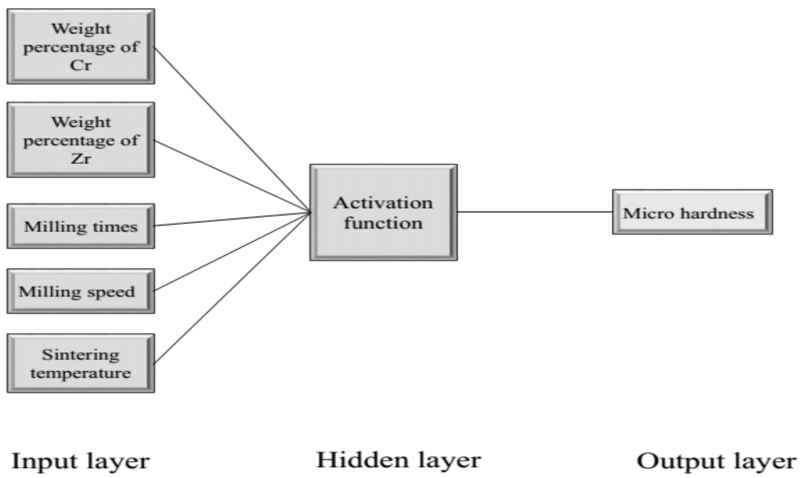

Fig. 4- Schematically representation of feed-forward neural networks.

ANN structures have been shown in Fig. 5.

In this analysis, a step-by-step method employed on the optimum ANN sensitivity changing in input parameter, one at a time, at a constant rate. Various constant rates 5, 10, 15 and 20 are selected for changing input parameter. eq. 8 estimates the sensitivity of each parameter:

$$S_i = \frac{1}{N}\sum_{1}^{n}\left(\frac{\%\ \text{Change in output}}{\%\ \text{Change in input}}\right) \times 100 \qquad \text{(eq. 8)}$$

In which, S_i (%) is sensitivity level of each input parameters and N is the total number of datasets, i.e., N=78.

Figure 6 represents the changes in hardness as a function of every input variable. As shown, the amounts of alloying elements (i.e., Cr and Zr) with direct positive effects are the most effecting parameters on the hardness of products. This can be related to the induction of severe distortion in Cu matrix during the solid solution preparation by mechanical alloying. Milling time and vial speed with dual effect situated at the later sequence on the hardness of products. This can be related to two opposite effects of these parameters during of milling. From one hand, by increasing of these parameter, the amount of transferred energy to the powders enhanced dramatically and as a consequence, preparation of finer grains with

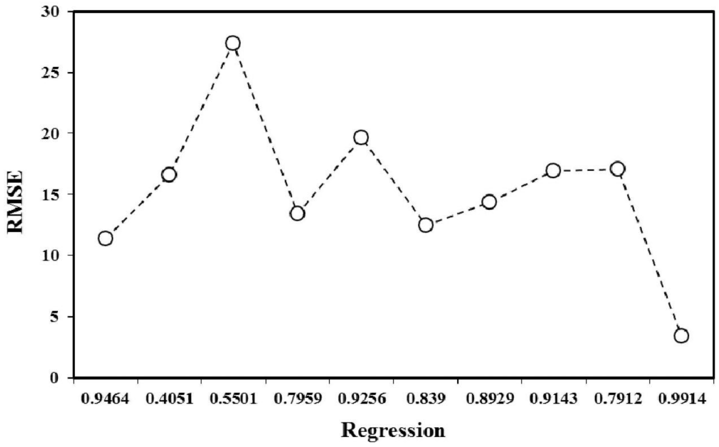

Fig. 5- RMSE values for different ANN structures.

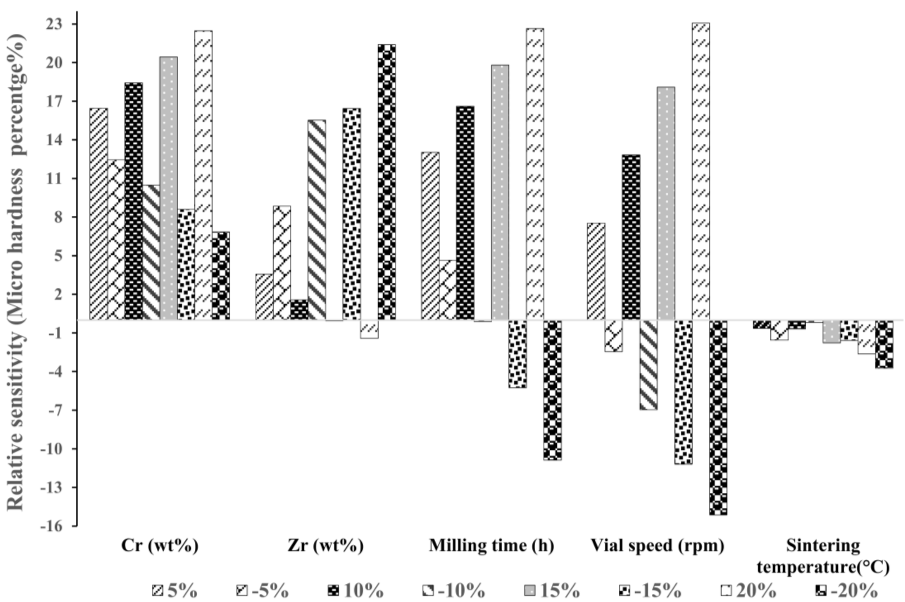

Fig. 6- The significance of input variables in micro hardness of Cu–Cr–Zr alloys.

higher hardness is encouraged and from the other hand, at higher milling times and vial speed, the possibility of products crystallization is increased due to the higher temperature increasing during the milling, consequently decreased the hardness of products. It was necessary to note that the sintering temperature showed the lowest negative effect on the hardness due the possibility of grain growth at higher temperature. According to the sensitivity analysis, the lowest effect of sintering temperature is an evident that the presence of alloying elements can be effectively decreased the possibility of grain growth at higher temperatures.

According to Literatures, various nanostructur materials are produced after initial crystallizaton of totally or partially amorhous precursores by controlling of the nucleation and growth process during the heating [31, 32]. In many cases, slowly diffiusing elements (e.g, substitual alloying element of Zr and Cr in present study) are added to control the graine growth and prepare finer distribution of grain size [33]. In the presence of such large substituational alloying elements, surface instability as a consequence of lower grain size is not occored during the crystallization and the concentration profile of these element around the growing crystallites affect both nucleation and growth of neighber graines, i.e., impingement effect [34, 35]. The solubility of alloying element (especially Cr) in Cu matrix is about zero and due to impingment effect of such element during the sintering the effect of temperature on the average particle size of Cu-Cr-Zr is lower than the other parmeters.

In this study, GA with a single point crossover and roulette wheel selection has been used. Every individual is prepared using a fitness function (i.e., the output of ANN model). The initial population is fixed to 650 generation size equal 100, crossover probability is 0.9 and mutation = 0.1. Typically, the optimum condition for preparation of Cu-Cr-Zr alloys is proposed as the combination of weight percentage of Cr and Zr as alloying element, milling times, milling speed and sintering temperature to

Table 4- Proposed optimum condition by GA

Matrix		Cu
Reinforcement	Cr Wt.%	3.4- 3.6
	Zr Wt.%	1-1.5
Milling Time (h)	71-81	
Burn-Rising Point (BRP)	10	
Vial Speed (rpm)	255-291	
Sintering Temperature (°C)	590-530	
Sintering Time (min)	30	

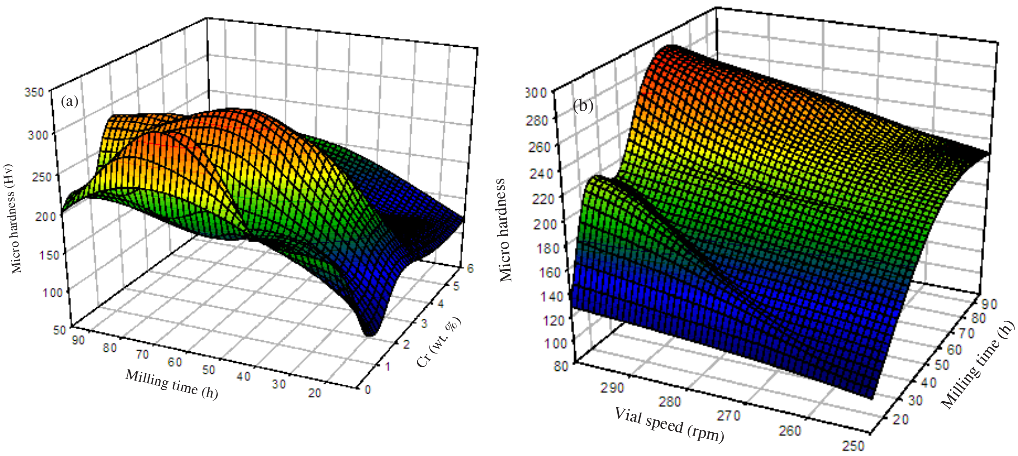

Fig. 7- (a) 3D profiles of micro hardness dependency of Cu–Cr–Zr alloys versus milling time and Cr (wt.%) at constant BPR=10:1; RPM=255; Sintering time= 30min, Sintering temperature= 590°C and Zr (wt%)=1.5%. (b) 3D profiles of micro hardness dependency of Cu–Cr–Zr alloys versus vial speed and milling time at constant Zr (wt%)=1.5%; Cr(wt%)=3(wt.%); Burning Through Point (BTP)= 10:1; rpm: 300; Sintering time: 30 min and Sintering temperature= 590°C.

enhance the micro hardness of proposed alloy to 310 micro vikers. To validate the simulation results, various configurations are checked as shown in table 4. The results confirmed that the optimum fitness function belong to trials number 71 and close to the condition predicted by GA and as consequence proved the validity of simulation results. According to the sensitivity analysis, Cr (wt.%), milling time and speed with positive effect are the most effective parameters on micro hardness of products. This can be related to the effect of these parameters for decreasing of particle size as well as barrier effect of alloying elements for dislocation movement. As shown in Fig. 7, the micro hardness does not continuously increase or decrease with any other parameters. This trend is a consequence of mutually effect of practical parameters on micro harness of products, e.g., two opposite effects of milling time and vial speed on micro hardness.

4. Conclusions

In this study combination of GA-ANN employed to determine the optimum condition for micro hardness maximization of Cu-Cr-Zr alloy prepared by mechanical alloying. The results can be categorized ad following:

ANN model with 30 and 15 neurons in hidden layers 1 and 2, respectively is a useful method for the prediction of hardness of Cu-Cr-Zr alloys fabricated by mechanical alloying;

The combined GA–ANN algorithm is an effective model for optimization of mechanical alloying parameters, leading to the maximum hardness in Cu-Cr-Zr alloys;

Sensitivity analysis shows that the alloying elements (Cr, Zr), vial speed and milling time are sintering temperature are the most important parameters on the hardness of products.

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