Grain size calculation of Cu-Zn alloys using genetic programming; an alternative for Scherer's formula

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Genetic Programming (GP) was used as a new method for formulation of grain size of electrodeposited $Cu_{1-x}Zn_x$ alloys as a function of Zinc and Copper content both electrolyte and the alloy films produced by electrodeposition technique. To predict grain size 48 different expression models were conducted. Each model differs from the other with their linking function, number of genes, head size, and chromosomes. To generate databases for the new grain size formulations, testing and training sets in total of 134 samples were selected at different Zn and Cu ratios of components. The testing and training sets consisted of randomly selected 106 and 28 for the proposed models. 6 different input parameters were selected as d-spacing spacing, Zn and Cu % content in the electrolyte and thin films and the test FWHM of the thin films obtained by XRD results. The output parameter was grain size of the electrodeposited Cu-Zn alloys. All results in the models indicated an applicable performance for predicting grain size of the alloys and found reliable. The predicted model showed that all of the input parameters effected on the resulting grain size.

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1. Introduction

There are a lot of new alloys in the technological world produced for different application. Shape Memory Alloys (SMA's) are a particular type of material that which changes its shape due to change in temperature. Beside these properties Cu-Zn alloys shows shape memory effect (SME). SMA's have remarkable properties such as SME and pseudoelasticity which is ally with the specific way the phase transformation occurs. Cu–Zn alloys have been extensively used in scientific applications and industry, owing to their attractive properties such as adhesion to steel, protection against corrosion, pipe and aircraft hydraulic couplings, various actuators in electric appliances, electrical connectors, microelectromechanical systems, automobile applications, antennae for cellular phones valves in fire-safety devices, robotic muscles, and surgical tools and biomedical implant materials, and decorative property [1- 6].

The preparation techniques are closely related with the structure of produced heterogeneous alloys [7-10]. Electrodeposition is a main technology for the deposition of metallic alloys. It is a low cost application and an alternative method to vacuum required systems such as sputtering, evaporation, Molecular Beam Epitaxy. With this method multilayer and immiscible metal combinations can be produced by control of the deposition variables such as pH, temperature, concentration, voltage and deposition current. Therefore, these processes have been developed for a wide range of applications, such as protective coatings, electronic industry and metal and metallic alloy film preparation in many technological processes.

The electrodeposition is affected by different deposition parameters [11-16]. The grain size of a deposited film can be altered by varying the composition of the electrolyte under given conditions. Dudin et al. [17] illustrated that the grain size of nickel coatings decreased from 70 nm to 40 nm by increasing the current density from 1 to 30 $mA.dm^{-2}$. Garcia et al. showed that the changing chemical composition of the coatings was changed the grain sizes of the Cu-Zn alloys [18].

Artificial neural networks, systems, fuzzy logic, adaptive neuro-fuzzy interfacial and GP are common Soft computing techniques are preferred if the numbers of the accessible data are appropriate. In our previous work [19], GP was used for the formulations of magnetoresistance and electrical resistivity properties of electrodeposited Cu-Co-Ni alloys. In another study, empirical formulations were proposed by applying the GP for prediction of electrical resistivity of Zn-Fe alloys [33]. There are a lot of papers about the applications of GEP in the literature for different engineering problems. Cevik and Guzelbey [20] used the GEP and predicted the ultimate strength of metal plates in compression. Eskil and Kanca [21] tried to develop a GP based formulation about the effect of

changing composition and heat treatments on the martensite start temperature of FeMnSi shape memory alloys. Nazari and Abdinejad predicted a new formulation for Charpy impact energy of laminated Al/SiCP nanocomposites [22]. In our latest study, empirical formulations were proposed by applying the NN and GEP for prediction of electrical resistivity of Zn-Fe alloys. These formulations were compared with each other [23].

The crystallographic texture, grain morphology, and grain size properties of thin films affected from the grain structure of the materials. Grain morphology and size is related to physical properties of thin film and the behavior of metals influenced with the grain size. Grain sizes in the electrodeposited materials are usually determined via direct observations under high magnified electron microscopy, or by estimations from the X-ray diffraction data. Generally, in the literature the grain size (D) was estimated using Scherer's formula [24].

$$D = \frac{0.9\,\lambda}{\beta \cos\theta}$$

where β the full width at half maximum of the peak in radians and θ the Bragg angle of the X-ray diffraction peak, and λ is the wavelength of X-ray used (1.5400 Å).

The above mentioned mathematical model was derived from physical descriptions. This model usually is computationally unclear, mathematically complex, and requires detailed knowledge of the XRD process. Therefore, alternative methods for designation of Scherer's formula by using available process data and enlarging it to a usable mathematical model, which can be applied simply on unavailable data, are so worthwhile. Genetic programming (GP), generally speaking genetic algorithm (GA), can be used in wide range of applications. Thus, to the best of our knowledge, there is no work in the field of modeling of grain size estimation by means of GP for the Cu-Zn alloys.

It is very useful formula to calculate the grain size practically with XRD data but all the physical properties vary with composition of film and electrolyte, and d-spacing an important another factor. The literature is lack of a clear formulation for estimating the grain size of electrodeposited alloys related to bath and film component. For this purpose, in this work, GEP was preferred to predict and present suitable formulation of grain size of electrodeposited Cu-Zn alloys. It would be useful to have a quantitative relationship between bath and film composition, FWHM, and grain size of Cu-Zn alloys. Hundred and thirty four input-target data were gathered from the previous work done in our laboratory, randomly selected and divided into 106 and 28 data sets and then were respectively trained and tested by the proposed models. The d-spacing of the samples, and FWHM values gained from the XRD results, weight percentages of zinc and iron in the film and in the electrolyte were considered as 6 independent input parameters.

2. Experimental details

Electrodeposition was realized in a conventional glassy cell in nonstirred and nonearrated conditions. The electrolyte composition and experimental parameters for the Cu-Zn films are reported in Table 1. The solutions were prepared using double distilled water and the chemicals are of high purity.

Aluminum plates were used as substrate and before deposition; the samples were polished, degreased, and activated by dipping in a 1 M NaOH with surfactant at 70 ^{o}C during 5 min and finally rinsed with the twice distilled water (18 MΩ cm) and dried with fresh air. The effects of bath composition on grain size behavior were investigated.

A Rigaku diffractometer was used to analyze the crystallographic structure of alloys. The XRD was operated at 30 kV and 30 mA with CuKα radiation. Energy Dispersive X-Ray Spectrometer (EDX) was used to determine the elemental composition of the films.

Table 1. Bath conditions of the $Cu_{1-x}Zn_x$ alloys.

Film No	Thin Films	Electrolyte in materials			pH	Current (mA)	Time (Min.)	Temperature °C	Aluminum Substrate area (cm^2)
		$CuSO_4.5H_2O$ (Mol/lt)	$ZnSO_4.7H2O$ (Mol/lt)	$Na_3C_6H_5O_7$ (Mol/lt)					
1	$Cu_{26}Zn_{74}$	0,06	0,2	0,5	5,8	60	60	20	3,8
2	$Cu_{37}Zn_{63}$	0,08	0,2	0,5	5,8	60	60	20	3,8
3	$Cu_{52}Zn_{48}$	0,1	0,2	0,5	5,8	60	60	20	3,8
4	$Cu_{62}Zn_{38}$	0,06	0,1	0,5	5,8	60	60	20	3,8
5	$Cu_{25}Zn_{75}$	0,06	0,3	0,5	5,8	60	60	20	3,8
6	$Cu_{21}Zn_{79}$	0,06	0,4	0,5	5,8	60	60	20	3,8
7	$Cu_{38}Zn_{62}$	0,06	0,2	0,3	5,8	60	60	20	3,8
8	$Cu_{26}Zn_{74}$	0,06	0,2	0,5	5,8	60	60	20	3,8
9	Cu_6Zn_{94}	0,06	0,2	0,7	5,8	60	60	20	3,8
10	$Cu_{13}Zn_{87}$	0,06	0,2	0,9	5,8	60	60	20	3,8
11	$Cu_{86}Zn_{14}$	0,08	0,2	0,5	5,8	10	40	20	3,8
12	$Cu_{66}Zn_{34}$	0,08	0,2	0,5	5,8	20	40	20	3,8
13	$Cu_{48}Zn_{52}$	0,08	0,2	0,5	5,8	30	40	20	3,8
14	$Cu_{47}Zn_{53}$	0,08	0,2	0,5	5,8	40	40	20	3,8
15	$Cu_{43}Zn_{57}$	0,08	0,2	0,5	5,8	60	40	20	3,8
16	$Cu_{17}Zn_{83}$	0,08	0,2	0,5	5,8	80	40	20	3,8
17	$Cu_{25}Zn_{75}$	0,08	0,2	0,5	5,8	100	40	20	3,8
18	$Cu_{13}Zn_{87}$	0,06	0,2	0,5	5,8	60	20	20	3,8
19	$Cu_{12}Zn_{88}$	0,06	0,2	0,5	5,8	60	30	20	3,8
20	Cu_5Zn_{95}	0,06	0,2	0,5	5,8	60	40	20	3,8
21	$Cu_{17}Zn_{83}$	0,06	0,2	0,5	5,8	60	50	20	3,8
22	Cu_7Zn_{93}	0,06	0,2	0,5	5,8	60	60	20	3,8

3. Genetic Programming

Genetic programming (GP) firstly proposed by Koza [25] takes its power from biological natural selection system and automatically solves problems using computer. The GP initialize a population and compound the random members known as chromosomes. Then, the fitness of each chromosome is evaluated with respect to a final amount. GP creates equal and unequal computer programs, which comprise of several variable sets and mathematical operators as the solution. The system can be comprised of function calls (ex, sqrt, x, sin, cos, tan, log, ln, power) and mathematical operations (-, x, /, +). Each function implicitly includes an assignment to a variable, which facilitates the use of multiple program outputs in GP, those side effects must be incorporated clearly [26].

The GP is capable of simultaneously solving a problem and evolving the architecture of the overall program. In this work, empirical formulas were used for prediction of grain size of electrochemically deposited $Cu_{1-x}Zn_x$ alloys. The aim of genetic programming is to find a program that well matches with the experimental results.

This side of program is very important for catch the nearest solution. GP creates the first population randomly from the previously defined space. GP gives a program as an output to the user [27].

3.1. Gene Expression Programming

Ferriera [28] was invented the Gene-Expression Programming (GEP) as a natural development of genetic algorithms and GP. 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. Chromosomes and expression trees are the two main parameters of GEP. Translation (The process of information decoding) is based on a set of rules. The genetic code is presents a dense link between the chromosome and the function. The program uses the languages of expression trees and the genes. This gives an advantage to the user to assume exactly the phenotype via Karva notation [29].

Table 2. The variables used in model construction with GEP

Code	Input variable	Range	Code	Output variable	Range
d0	d	1.2031-2.7251	D.V	G	33.125-771.125 (n**m**)
d1	F	0.11-1.148			
d2	E_{Cu}	13.04-37.5			
d3	E_{Zn}	62.50-86.96			
d4	F_{Cu}	4.76-85.56			
d5	F_{Zn}	14.45-95.24			

*Element compositions of E_{Cu}, E_{Zn}, F_{Cu} and F_{Zn} are presented as wt %.

4. Application of genetic programming

The data used for the modeling of the grain size of $Cu_{1-x}Zn_x$ alloys was obtained from our experimental results. The major task herein is to define the hidden function connecting the input variables (d0, d1, d2, d3, d4 and d5) and output D.V. Following equations show the empirical models as a function of the experimental conditions.

$$D.V = f(d0, d1, d2, d3, d4 \text{ and } d5) \quad (1)$$

$$G = f(d, F, E_{Cu}, E_{Zn}, F_{Cu} \text{ ve } F_{Zn}) \quad (2)$$

The formulas obtained by GEP will be used for estimating the relationship between film components and grain size characteristic of $Cu_{1-x}Zn_x$ alloys. The variables of the GEP models were presented in Table 2.

The database is divided into two sets; test and training. To measure their generalization capabilities and to test the proposed models, the formulations were improved based on the former while the latter was employed [21]. The training and testing sets created from randomly selected 106 and 28 mixtures of all 134 alloys, respectively. It must be kept in mind that the proposed empirical equations are valid for the ranges of training and testing set given in Table 3 and Table 4, respectively. To show the performance of the models belonging the program, parameters of the GEP were presciently so as to predict the grain size of Cu-Zn alloys.

Table 3. Results of GEP formulations versus experimental training results

No	Thin Films	d Å	F FWHM	E_{Cu} wt.%	E_{Zn} wt.%	F_{Cu} wt.%	F_{Zn} wt.%	G Experimental results Nm	RsG Gep	$\frac{G}{R_s G}$
1	$Cu_{26}Zn_{74}$	2.5691	0.405	23.08	76.92	25.61	74.39	83.913	89.312	0.940
2	$Cu_{26}Zn_{74}$	2.0924	0.382	23.08	76.92	25.61	74.39	100.102	96.126	1.041
3	$Cu_{26}Zn_{74}$	1.8917	0.393	23.08	76.92	25.61	74.39	106.117	100.992	1.051
4	$Cu_{26}Zn_{74}$	1.2802	0.131	23.08	76.92	25.61	74.39	771.124	782.974	0.985
5	$Cu_{26}Zn_{74}$	1.2066	0.590	23.08	76.92	25.61	74.39	255.524	259.455	0.985
6	$Cu_{37}Zn_{63}$	2.1369	0.526	28.57	71.43	36.36	63.46	71.601	70.520	1.015
7	$Cu_{37}Zn_{63}$	2.0906	0.579	28.57	71.43	36.36	63.46	66.086	64.875	1.019
8	$Cu_{37}Zn_{63}$	1.2084	0.648	28.57	71.43	36.36	63.46	229.650	244.956	0.938
9	$Cu_{52}Zn_{48}$	2.1346	0.541	33.33	66.67	52.34	47.66	69.669	69.387	1.004
10	$Cu_{52}Zn_{48}$	2.0924	0.523	33.33	66.67	52.34	47.66	73.116	71.997	1.016
11	$Cu_{52}Zn_{48}$	1.8504	0.544	33.33	66.67	52.34	47.66	78.421	76.420	1.026
12	$Cu_{52}Zn_{48}$	1.3030	0.562	33.33	66.67	52.34	47.66	164.787	187.060	0.881
13	$Cu_{52}Zn_{48}$	1.2031	0.374	33.33	66.67	52.34	47.66	413.855	395.181	1.047
14	$Cu_{62}Zn_{38}$	2.1326	0.406	37.50	62.50	62.14	37.86	92.896	88.506	1.050
15	$Cu_{62}Zn_{38}$	2.0880	0.430	37.50	62.50	62.14	37.86	89.070	85.096	1.047
16	$Cu_{62}Zn_{38}$	1.8386	0.400	37.50	62.50	62.14	37.86	107.382	101.252	1.061
17	$Cu_{62}Zn_{38}$	1.3066	0.188	37.50	62.50	62.14	37.86	486.372	458.324	1.061
18	$Cu_{62}Zn_{38}$	1.2066	0.540	37.50	62.50	62.14	37.86	279.236	293.420	0.952

No	Thin Films	d Å	F FWHM	E_{Cu} wt.%	E_{Zn} wt.%	F_{Cu} wt.%	F_{Zn} wt.%	G Experimental results Nm	RsG Gep	$\frac{G}{R_s G}$
19	$Cu_{25}Zn_{75}$	2.5726	0.537	16.67	83.33	25.18	74.82	63.249	62.359	1.014
20	$Cu_{25}Zn_{75}$	2.0900	0.401	16.67	83.33	25.18	74.82	95.442	85.767	1.113
21	$Cu_{25}Zn_{75}$	1.8900	0.342	16.67	83.33	25.18	74.82	122.051	112.963	1.080
22	$Cu_{25}Zn_{75}$	1.8136	0.318	16.67	83.33	25.18	74.82	137.129	129.585	1.058
23	$Cu_{25}Zn_{75}$	1.5869	0.299	16.67	83.33	25.18	74.82	176.310	175.777	1.003
24	$Cu_{25}Zn_{75}$	1.4795	0.349	16.67	83.33	25.18	74.82	174.443	175.945	0.991
25	$Cu_{25}Zn_{75}$	1.3723	0.420	16.67	83.33	25.18	74.82	179.465	183.933	0.976
26	$Cu_{25}Zn_{75}$	1.3081	0.186	16.67	83.33	25.18	74.82	488.989	524.744	0.932
27	$Cu_{25}Zn_{75}$	1.2528	0.371	16.67	83.33	25.18	74.82	308.048	315.136	0.978
28	$Cu_{25}Zn_{75}$	1.2097	0.426	16.67	83.33	25.18	74.82	346.034	333.529	1.037
29	$Cu_{21}Zn_{79}$	1.8900	0.358	13.04	86.96	20.67	79.33	116.596	99.364	1.173
30	$Cu_{21}Zn_{79}$	1.8211	0.180	13.04	86.96	20.67	79.33	241.154	248.834	0.969
31	$Cu_{21}Zn_{79}$	1.4793	0.297	13.04	86.96	20.67	79.33	205.047	204.816	1.001
32	$Cu_{21}Zn_{79}$	1.2077	0.563	13.04	86.96	20.67	79.33	265.562	241.411	1.100
33	$Cu_{38}Zn_{62}$	2.5646	0.417	23.08	76.92	38.31	61.69	81.562	86.921	0.938
34	$Cu_{38}Zn_{62}$	2.3661	0.285	23.08	76.92	38.31	61.69	124.114	125.130	0.992
35	$Cu_{38}Zn_{62}$	2.0944	0.336	23.08	76.92	38.31	61.69	113.727	109.870	1.035
36	$Cu_{38}Zn_{62}$	1.8153	0.261	23.08	76.92	38.31	61.69	166.908	163.841	1.019
37	$Cu_{38}Zn_{62}$	1.7393	0.230	23.08	76.92	38.31	61.69	199.419	198.716	1.004
38	$Cu_{38}Zn_{62}$	1.5887	0.298	23.08	76.92	38.31	61.69	176.545	178.125	0.991
39	$Cu_{38}Zn_{62}$	1.4824	0.411	23.08	76.92	38.31	61.69	147.453	151.492	0.973
40	$Cu_{38}Zn_{62}$	1.2532	0.382	23.08	76.92	38.31	61.69	298.597	310.681	0.961
41	$Cu_{38}Zn_{62}$	1.2096	0.468	23.08	76.92	38.31	61.69	315.179	313.739	1.005
42	$Cu_{26}Zn_{74}$	2.5718	0.414	23.08	76.92	25.61	74.39	82.052	87.565	0.937
43	$Cu_{26}Zn_{74}$	2.3864	0.298	23.08	76.92	25.61	74.39	118.160	118.836	0.994
44	$Cu_{26}Zn_{74}$	2.0923	0.408	23.08	76.92	25.61	74.39	93.728	89.737	1.044
45	$Cu_{26}Zn_{74}$	1.8923	0.407	23.08	76.92	25.61	74.39	102.437	97.245	1.053
46	$Cu_{26}Zn_{74}$	1.4775	0.478	23.08	76.92	25.61	74.39	127.772	131.495	0.972
47	$Cu_{26}Zn_{74}$	1.2793	0.219	23.08	76.92	25.61	74.39	463.040	477.698	0.969
48	$Cu_{6}Zn_{94}$	2.7251	0.132	23.08	76.92	6.15	93.85	251.335	251.304	1.000
49	$Cu_{6}Zn_{94}$	2.5573	0.351	23.08	76.92	6.15	93.85	97.021	101.377	0.957
50	$Cu_{6}Zn_{94}$	2.3873	0.294	23.08	76.92	6.15	93.85	119.747	119.491	1.002
51	$Cu_{6}Zn_{94}$	2.1462	0.174	23.08	76.92	6.15	93.85	215.793	210.144	1.027
52	$Cu_{6}Zn_{94}$	1.8096	0.125	23.08	76.92	6.15	93.85	349.745	348.154	1.005
53	$Cu_{6}Zn_{94}$	1.6411	0.183	23.08	76.92	6.15	93.85	272.309	275.298	0.989
54	$Cu_{6}Zn_{94}$	1.5974	0.290	23.08	76.92	6.15	93.85	179.692	181.672	0.989
55	$Cu_{6}Zn_{94}$	1.4807	0.573	23.08	76.92	6.15	93.85	106.047	109.497	0.968
56	$Cu_{6}Zn_{94}$	1.2073	0.586	23.08	76.92	6.15	93.85	256.034	261.399	0.979
57	$Cu_{13}Zn_{87}$	2.3961	0.271	23.08	76.92	13.01	86.99	129.659	129.602	1.000
58	$Cu_{13}Zn_{87}$	2.3251	0.125	23.08	76.92	13.01	86.99	285.716	283.066	1.009
59	$Cu_{13}Zn_{87}$	2.0948	0.385	23.08	76.92	13.01	86.99	99.241	95.521	1.039
60	$Cu_{13}Zn_{87}$	1.9069	0.425	23.08	76.92	13.01	86.99	97.362	92.566	1.052

No	Thin Films	d Å	F FWHM	E_{Cu} wt.%	E_{Zn} wt.%	F_{Cu} wt.%	F_{Zn} wt.%	G Experimental results Nm	RsG Gep	$\frac{G}{R_s G}$
61	$Cu_{13}Zn_{87}$	1.5979	0.306	23.08	76.92	13.01	86.99	170.193	171.670	0.991
62	$Cu_{13}Zn_{87}$	1.4836	0.567	23.08	76.92	13.01	86.99	106.676	109.596	0.973
63	$Cu_{13}Zn_{87}$	1.2299	0.293	23.08	76.92	13.01	86.99	441.458	439.989	1.003
64	$Cu_{13}Zn_{87}$	1.2104	0.578	23.08	76.92	13.01	86.99	253.885	259.970	0.977
65	$Cu_{86}Zn_{14}$	2.0859	0.706	28.57	71.43	85.56	14.45	54.290	53.367	1.017
66	$Cu_{66}Zn_{34}$	2.1083	1.148	28.57	71.43	65.55	34.45	33.125	33.483	0.989
67	$Cu_{66}Zn_{34}$	1.8677	0.110	28.57	71.43	65.55	34.45	384.054	379.853	1.011
68	$Cu_{48}Zn_{52}$	2.0903	0.504	28.57	71.43	47.43	52.58	75.928	73.809	1.029
69	$Cu_{47}Zn_{53}$	2.1337	0.387	28.57	71.43	47.31	52.69	97.423	94.398	1.032
70	$Cu_{47}Zn_{53}$	2.0948	0.582	28.57	71.43	47.31	52.69	65.647	64.244	1.022
71	$Cu_{47}Zn_{53}$	1.2084	0.462	28.57	71.43	47.31	52.69	322.048	323.362	0.996
72	$Cu_{43}Zn_{57}$	2.5653	0.456	28.57	71.43	43.33	56.67	74.577	81.355	0.917
73	$Cu_{43}Zn_{57}$	1.4797	0.499	28.57	71.43	43.33	56.67	121.968	127.955	0.953
74	$Cu_{43}Zn_{57}$	1.2079	0.554	28.57	71.43	43.33	56.67	269.678	278.889	0.967
75	$Cu_{17}Zn_{83}$	2.3961	0.278	28.57	71.43	16.62	83.39	126.392	124.278	1.017
76	$Cu_{17}Zn_{83}$	2.1541	0.154	28.57	71.43	16.62	83.39	243.198	228.829	1.063
77	$Cu_{17}Zn_{83}$	2.0973	0.402	28.57	71.43	16.62	83.39	94.959	92.043	1.032
78	$Cu_{17}Zn_{83}$	1.8980	0.231	28.57	71.43	16.62	83.39	179.944	171.071	1.052
79	$Cu_{17}Zn_{83}$	1.4838	0.418	28.57	71.43	16.62	83.39	144.653	150.955	0.958
80	$Cu_{17}Zn_{83}$	1.3820	0.230	28.57	71.43	16.62	83.39	320.084	329.126	0.973
81	$Cu_{17}Zn_{83}$	1.2818	0.189	28.57	71.43	16.62	83.39	531.061	527.474	1.007
82	$Cu_{17}Zn_{83}$	1.2145	0.985	28.57	71.43	16.62	83.39	144.764	170.456	0.849
83	$Cu_{25}Zn_{75}$	2.5540	0.564	28.57	71.43	24.87	75.13	60.414	68.550	0.881
84	$Cu_{25}Zn_{75}$	2.3871	0.412	28.57	71.43	24.87	75.13	85.453	87.312	0.979
85	$Cu_{25}Zn_{75}$	2.3208	0.179	28.57	71.43	24.87	75.13	199.729	191.124	1.045
86	$Cu_{25}Zn_{75}$	2.0947	0.401	28.57	71.43	24.87	75.13	95.283	92.194	1.034
87	$Cu_{25}Zn_{75}$	1.8342	0.262	28.57	71.43	24.87	75.13	164.374	157.372	1.044
88	$Cu_{25}Zn_{75}$	1.5974	0.155	28.57	71.43	24.87	75.13	336.188	330.307	1.018
89	$Cu_{25}Zn_{75}$	1.4798	0.519	28.57	71.43	24.87	75.13	117.252	123.872	0.947
90	$Cu_{25}Zn_{75}$	1.2849	0.392	28.57	71.43	24.87	75.13	252.874	269.913	0.937
91	$Cu_{13}Zn_{87}$	2.0902	0.429	23.08	76.92	13.31	86.70	89.207	85.552	1.043
92	$Cu_{13}Zn_{87}$	1.4783	0.229	23.08	76.92	13.31	86.70	266.386	277.293	0.961
93	$Cu_{13}Zn_{87}$	1.2079	0.328	23.08	76.92	13.31	86.70	455.450	438.622	1.038
94	$Cu_{12}Zn_{88}$	2.0902	0.523	23.08	76.92	11.75	88.26	73.174	69.780	1.049
95	$Cu_{12}Zn_{88}$	1.2053	0.539	23.08	76.92	11.75	88.26	282.348	284.047	0.994
96	$Cu_{5}Zn_{95}$	2.0764	0.370	23.08	76.92	4.76	95.24	103.949	100.158	1.038
97	$Cu_{5}Zn_{95}$	1.6784	0.330	23.08	76.92	4.76	95.24	145.952	144.529	1.010
98	$Cu_{5}Zn_{95}$	1.4723	0.411	23.08	76.92	4.76	95.24	149.878	156.171	0.960
99	$Cu_{5}Zn_{95}$	1.2047	0.489	23.08	76.92	4.76	95.24	312.624	311.213	1.005
100	$Cu_{17}Zn_{83}$	2.5955	0.242	23.08	76.92	17.26	82.74	139.814	144.193	0.970
101	$Cu_{17}Zn_{83}$	2.3842	0.386	23.08	76.92	17.26	82.74	91.268	92.156	0.990
102	$Cu_{17}Zn_{83}$	2.0993	0.525	23.08	76.92	17.26	82.74	72.660	69.133	1.051

No	Thin Films	d Å	F FWHM	E_{Cu} wt.%	E_{Zn} wt.%	F_{Cu} wt.%	F_{Zn} wt.%	G Experimental results Nm	RsG Gep	$\frac{G}{R_s G}$
103	$Cu_{17}Zn_{83}$	1.3767	0.343	23.08	76.92	17.26	82.74	217.381	231.358	0.940
104	$Cu_{17}Zn_{83}$	1.2097	0.422	23.08	76.92	17.26	82.74	349.472	345.590	1.011
105	$Cu_{7}Zn_{93}$	1.4862	0.374	23.08	76.92	7.08	92.92	161.076	167.091	0.964
106	$Cu_{7}Zn_{93}$	1.2105	0.344	23.08	76.92	7.08	92.92	426.316	415.051	1.027

Table 4. Results of GEP formulations versus experimental testing results

No	Thin Films	d Å	F (FWHM	E_{Cu} wt.%	E_{Zn} wt.%	F_{Cu} wt.%	F_{Zn} wt.%	G xperimental results Nm	RsG Gep	$\frac{D}{R_s G}$
1	$Cu_{26}Zn_{74}$	1.4794	0.481	23.08	76.92	25.61	74.39	126.583	130.154	0.973
2	$Cu_{37}Zn_{63}$	1.4787	0.465	28.57	71.43	36.36	63.46	131.099	137.279	0.955
3	$Cu_{52}Zn_{48}$	1.8650	0.784	33.33	66.67	52.34	47.66	53.966	54.361	0.993
4	$Cu_{62}Zn_{38}$	1.4764	0.411	37.50	62.50	62.14	37.86	148.870	153.347	0.971
5	$Cu_{25}Zn_{75}$	2.3694	0.367	16.67	83.33	25.18	74.82	96.310	92.685	1.039
6	$Cu_{25}Zn_{75}$	1.7399	0.341	16.67	83.33	25.18	74.82	134.447	126.596	1.062
7	$Cu_{25}Zn_{75}$	1.2800	0.344	16.67	83.33	25.18	74.82	293.923	306.273	0.960
8	$Cu_{21}Zn_{79}$	2.0966	0.452	13.04	86.96	20.67	79.33	84.475	65.317	1.293
9	$Cu_{38}Zn_{62}$	1.8940	0.363	23.08	76.92	38.31	61.69	114.751	109.549	1.047
10	$Cu_{38}Zn_{62}$	1.2815	0.296	23.08	76.92	38.31	61.69	339.561	355.062	0.956
11	$Cu_{26}Zn_{74}$	2.1452	0.097	23.08	76.92	25.61	74.39	387.221	395.728	0.979
12	$Cu_{26}Zn_{74}$	1.2078	0.688	23.08	76.92	25.61	74.39	217.193	224.912	0.966
13	$Cu_{6}Zn_{94}$	2.0900	0.389	23.08	76.92	6.15	93.85	98.385	94.790	1.038
14	$Cu_{6}Zn_{94}$	1.3768	0.364	23.08	76.92	6.15	93.85	204.796	218.768	0.936
15	$Cu_{13}Zn_{87}$	2.1520	0.210	23.08	76.92	13.01	86.99	178.469	174.425	1.023
16	$Cu_{13}Zn_{87}$	1.3820	0.414	23.08	76.92	13.01	86.99	177.832	189.954	0.936
17	$Cu_{86}Zn_{14}$	2.0925	0.501	28.57	71.43	65.55	34.45	76.324	74.068	1.030
18	$Cu_{47}Zn_{53}$	1.2067	0.413	28.57	71.43	47.43	52.58	364.696	358.413	1.018
19	$Cu_{43}Zn_{57}$	2.0949	0.429	28.57	71.43	43.33	56.67	89.057	86.189	1.033
20	$Cu_{17}Zn_{83}$	2.5857	0.349	28.57	71.43	16.62	83.39	97.106	101.925	0.953
21	$Cu_{17}Zn_{83}$	1.6000	0.278	28.57	71.43	16.62	83.39	186.913	185.650	1.007
22	$Cu_{17}Zn_{83}$	1.2092	0.620	28.57	71.43	16.62	83.39	238.667	253.689	0.941
23	$Cu_{25}Zn_{75}$	1.8954	0.362	28.57	71.43	24.87	75.13	114.981	110.160	1.044
24	$Cu_{25}Zn_{75}$	1.2097	0.667	28.57	71.43	24.87	75.13	220.985	238.134	0.928
25	$Cu_{13}Zn_{87}$	2.4843	0.239	23.08	76.92	13.31	86.70	144.394	145.656	0.991
26	$Cu_{5}Zn_{95}$	2.8757	0.093	23.08	76.92	4.76	95.24	349.947	347.046	1.008
27	$Cu_{17}Zn_{83}$	1.4786	0.255	23.08	76.92	17.26	82.74	239.087	248.754	0.961
28	$Cu_{7}Zn_{93}$	2.0949	0.401	23.08	76.92	7.08	92.92	95.277	91.762	1.038

4.1 GEP formulations

Tool parameters, the list of function and all tried combinations obtained from the experimental results were given above. Table 5 and 6 presents the GEP parameters, and the list of function, respectively. All tried combinations are given in Table 7. To gain from the computational time, a subset of combinations is selected instinctively and performance of the GEP algorithm in predicting the grain size was determined. The optimal setting was used for the prediction of grain size with the equations below:

$$U_1 = \frac{(-3)\cdot F_{Cu}+d\cdot E_{Zn}}{E_{Cu}} - \frac{9\cdot E_{Zn}}{E_{Cu}} \quad (3)$$

$$U_2 = \frac{\frac{E_{Cu}}{\sqrt{d+F}\cdot(d^2-d)}}{\sqrt{F_{Zn}}} \quad (4)$$

$$U_3 = \frac{E_{Zn}}{F\cdot\left(4-\frac{4}{d}\right)} \quad (5)$$

$$U_4 = d^2\cdot\left(6+\frac{F_{Cu}}{(F\cdot E_{Cu})^2}\right) \quad (6)$$

$$U_5 = \frac{1}{d^8\cdot F}\cdot E_{Zn} \quad (7)$$

$$G = U_1 + U_2 + U_3 + U_4 + U_5 \quad (8)$$

$$G = \frac{(-3)\cdot F_{Cu}+d\cdot E_{Zn}}{E_{Cu}} - \frac{9\cdot E_{Zn}}{E_{Cu}} + \frac{\frac{E_{Cu}}{\sqrt{d+F}\cdot(d^2-d)}}{\sqrt{F_{Zn}}} + \frac{E_{Zn}}{F\cdot\left(4-\frac{4}{d}\right)} + d^2\cdot\left(6+\frac{F_{Cu}}{(F\cdot E_{Cu})^2}\right) + \frac{1}{d^8\cdot F}\cdot E_{Zn} \quad (9)$$

The functions generated for the best solutions by GEP algorithm to estimate the grain size predictions of electrodeposited alloys were presented in Equation 9.

Table 5. List of function set (p1)

Code	Function set (p1)
S1	+,-,*,/
S2	+,-,*,/,$\sqrt{x}$
S3	+,-,*,/,$\sqrt{x}$, x^2
S4	+,-,*,/,$\sqrt{x}$, x^2, x^3, $\sqrt[3]{x}$
S5	+,-,*,/,$\sqrt{x}$, x^2, ln(x), Sin(x)
S6	+,-,*,/,$\sqrt{x}$, x^2, x^3,ln(x), $\sqrt[3]{x}$
S7	+,-,*,/,$\sqrt{x}$, x^2, x^3, ln(x), $\sqrt[3]{x}$, Sin(x), Cos(x)
S8	+,-,*,/, x^2, x^3, exp(x), ln(x), Arctg(x)

Table 6. The best results obtained from the GEP tests

P1	P2	P3	P4	P5	P6	P7	R^2 error	
							Training data	Test data
S1	50	10	5	290481	Addition	RRSE	0,956	0,923
S1	50	8	4	336112	Multiplication	RRSE	0,983	0,973
S1	50	8	5	439983	Multiplication	rRRSE	0,988	0,985
S1	50	8	5	302059	Division	RRSE	0,965	0,777
S1	60	8	5	259622	Multiplication	rMSE	0,937	0,889
S1	70	8	5	347670	Addition	RRSE	0,963	0,912
S1	60	10	5	352105	Multiplication	rRMSE	0,991	0,982
S1	50	8	4	235082	Subtraction	RRSE	0,963	0,965
S1	60	8	5	300802	Addition	MAE	0,995	0,987
S1	60	8	5	380052	Multiplication	RAE	0,996	0,987
S2	50	10	5	323596	Addition	RRSE	0,948	0,851
S3	40	8	4	108918	Addition	RRSE	0,947	0,925
S3	50	8	5	549690	Addition	RRSE	0,984	0,961
S3	50	9	5	169887	Addition	MSE	0,996	0,991
S3	40	8	5	212519	Addition	rMSE	0,925	0,936
S3	50	8	5	303301	Addition	RRSE	0,958	0,960

P1	P2	P3	P4	P5	P6	P7	R^2 error	
							Training data	Test data
S3	50	10	5	220154	Addition	RMSE	0,968	0,961
S3	50	10	5	196669	Multiplication	RRSE	0,914	0,830
S3	50	8	5	549690	Addition	RRSE	0,984	0,961
S3	50	8	5	236282	Addition	RRSE	0,976	0,955
S3	50	8	5	484072	Addition	RRSE	0,984	0,961
S3	**50**	**10**	**5**	**472315**	**Addition**	**RRSE**	**0,995**	**0,993**
S3	50	8	5	303308	Addition	RRSE	0,994	0,980
S3	50	10	5	361196	Multiplication	RRSE	0,968	0,925
S3	50	10	5	753932	Multiplication	RRSE	0,924	0,729
S3	60	8	6	286703	Multiplication	MSE	0,995	0,986
S3	60	80	6	477545	Addition	rRMSE	0,964	0,972
S3	70	10	5	370301	Addition	rRMSE	0,995	0,992
S3	60	8	5	336112	Addition	rRAE	0,985	0,992
S3	30	8	3	314141	Addition	RRSE	0,971	0,950
S3	45	9	5	131866	Addition	RRSE	0,948	0,940
S4	50	10	5	183761	Multiplication	RRSE	0,966	0,940
S5	50	10	5	252260	Addition	RRSE	0,969	0,952
S5	50	10	5	297513	Multiplication	RRSE	0,969	0,930
S5	50	10	5	369003	Division	RRSE	0,982	0,882
S5	50	9	5	366344	Addition	rRMSE	0,995	0,991
S6	50	10	5	216259	Multiplication	RMSE	0,977	0,980
S6	50	8	5	322318	Multiplication	MAE	0,989	0,973
S6	50	10	5	203609	Multiplication	rMSE	0,988	0,972
S6	50	10	3	268345	Addition	MSE	0,983	0,973
S6	50	10	6	312070	Addition	MSE	0,981	0,952
S6	40	10	5	517679	Addition	RRSE	0,967	0,936
S7	50	10	6	272447	Multiplication	RRSE	0,991	0,978
S7	50	10	5	230711	Multiplication	RRSE	0,989	0,969
S7	50	8	5	406871	Addition	RRSE	0,995	0,993
S7	50	8	5	336112	Addition	rRRSE	0,956	0,970
S7	50	10	5	356846	Addition	RSE	0,993	0,965
S8	50	10	5	361196	Multiplication	RRSE	0,968	0,925

5. Results and Discussion

Experimental and calculated values via GEP formulations are compared for the grain size in Figure 1, 2. It was observed in Figs 1, 2 that the proposed GEP formulation for grain size of Cu-Zn alloys is able to follow closely the trend seen in the experimental data within test sets.

It was observed in Figure 1, 2 that the proposed model for the grain size provided consistent predictions for test data sets.

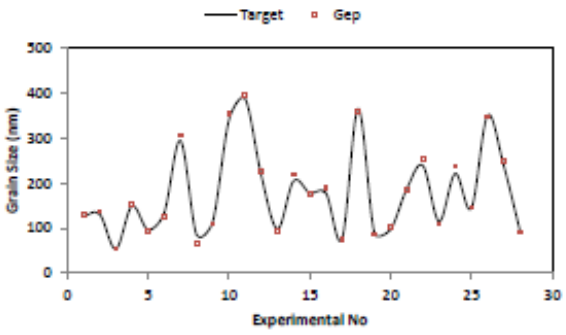

Fig.1 Testing evaluation of the GEP method for the grain size prediction as a function of experiment number

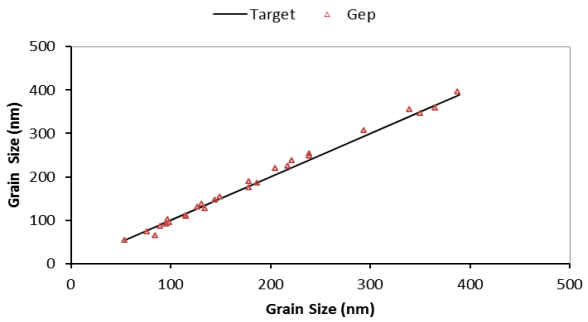

Fig.2 Testing evaluation of the GEP method for the grain size prediction

The figures showed clearly that there was a clear noticeably different between the predicted and the experimental values when the model was applied to the test set. However, this model conformed well to the experimental values in the train set.

Table 7 shows the statistical parameters of both test and training sets of these intelligence formulations, where R is the coefficient of correlation; RSE is the relative squared error, RRSE is the root relative squared error; RAE is the relative absolute error. As can be seen in Table 7, correlation coefficient of the train set of empirical model is higher than correlation coefficient of the testing set.

Table 7. GEP parameters used for proposed models and statistical

p1	Function set	+, -, *, /, $\sqrt{x}$, ln(x), x^2, x^3, $\sqrt[3]{x}$, Sin(x), Cos(x), Exp(x), Arctg(x)
p2	Chromosomes	50
p3	Head size	10
p4	Number of genes	5
p5	Number of generation	472315
p6	Linking function	Addition
p7	Fitness Function	RRSE
p8	Mutation rate	0.044
p9	Inversion rate	0.1
p10	One-point recombination rate	0.3
p11	Two-point recombination rate	0.3
p12	Gene recombination rate	0.1
p13	Gene transposition rate	0.1
p14	IS Transpositon	0,1
p15	RIS Transposition	0,1
P16	Independent variables	6
p17	Trainig samples	106
p18	Testing samples	28
p19	Training fitness	934.55
P20	Training R-square	0.9951
P21	Training correlation coefficient	0.9975
P22	Training RRSE	0.0700
P23	Training RSE	0.0049
P24	Training RAE	0.0619
P25	Testing fitness	913.90
P26	Testing R-square	0.9939
P27	Testing correlation coefficient	0.9969
P28	Testing RRSE	0.0942
P29	Testing RSE	0.0088
P30	Testing RAE	0.0929
P31	Maximum fitness	1000
P32	Constants per gene	1
P33	Data Type	İnteger

Metallic thin films used in a lot of technological area such as microelectronic devices and magnetic storage media. The grain size of deposits depends on the alloy composition and usually decreases with an increase in the dissolved component concentration. The physical properties (density, mechanical strength, electrical and thermal) of the metallic thin films are much more strongly influenced by such factors as average grain size, grain shape, grain size distribution, and crystallographic texture than in the case of mechanically thin films.

Grain size of the thin films influences from electrolyte composition thereby film composition, microstructure, d-spacing, and etc. The plots of the grain size changes due to d-spacing of the samples, FWHM values gained from the XRD results, weight percentages of zinc and iron in the film and in the electrolyte by using the GEP equation were showed so it can be seen that our equations give reliable results with literature. Different input values were taken as constant for every figure. In some figures both alloying element in the film and elemental concentration in the electrolyte were changed together because these values changes interdependent. To define the effects of alloying elements on grain size, only the input parameters which were tested changed from the beginning region to the end of the region given in Table 2 which was obtained from our own experiments.

Grain size for all the samples shows dispersion with electrolyte Cu content as shown in Fig. 3, and 4. It is obvious that grain size and Cu content are related with structure of the film. For this purpose a relationship between grain size and Cu content was researched. The predicted grain sizes of Cu–Zn alloy continuously decreases with increasing addition level of Cu concentration of holding with increase in addition level from 16 to 25% similar to that observed with experimentally obtained grain sizes. At higher addition levels, caused an increase in the grain size. When the addition level of copper content is increased, the number of nucleating particles added will increase. As the number of particles increase the inter-particle distance decreases.

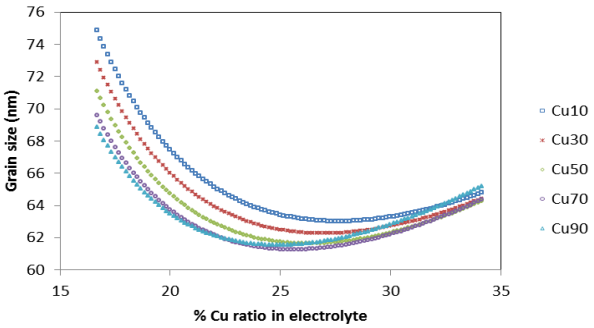

Fig.3 Variation of grain size as a function of Cu ratio in the film and electrolyte for the Cu_{1-x} Zn_x (x = 10–90) system at constant FWHM (0.6) value.

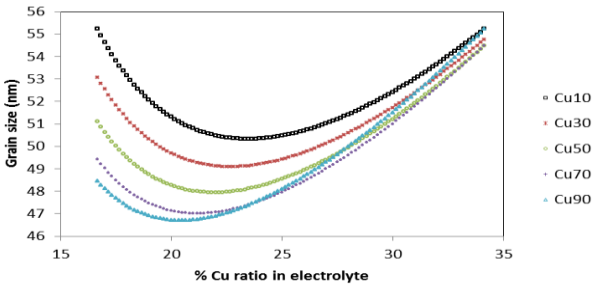

Fig.4 Effect of Cu ratio in the film and electrolyte for different copper content at the film and constant FWHM (0.75) value.

At large addition levels of the alloying element (Cu), it is possible that the inter-particle distance reduces. Thus, the fraction of particles that act as nucleating sites decreases with increasing addition level and thus the grain size reaches a more or less a saturation level. It is important to note that the GEP model is able to recognize the phenomenon. This expected result stem from typical metallic alloy behavior. It has been clearly shown that the GEP formulation is capable of giving well-matched values with literature [30, 31].

For changing d-spacing and FWHM values effect of Cu content in the film and electrolyte shown in Fig. 5. The more increase in upper level Cu ratio was not observed in this figure.

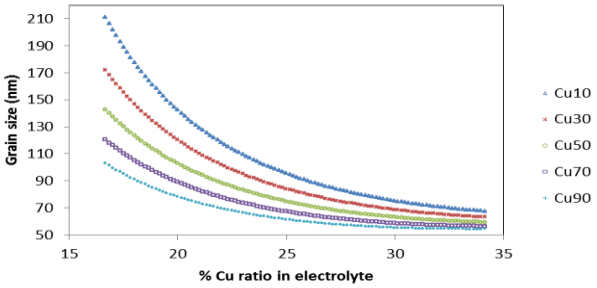

Fig.5 Effect of Cu ratio in the film and electrolyte variations on the grain size of electrodeposited Cu-Zn film predicted by GEP for changing d-spacing and FWHM values.

Fig 6 illustrates increase in grain size values obtained from GEP for the changing FWHM and electrolyte copper ratio. The grain size decreases with increasing copper ratio in the electrolyte. Our results, in accordance with international literature [32]

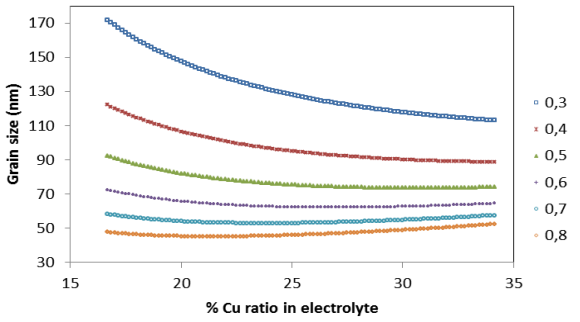

Fig.6 Effect of FWHM and electrolyte copper ratio variations on the grain size of electrodeposited Cu-Zn film predicted by GEP for the $Cu_{35}Zn_{65}$ film.

Fig. 7 and Fig.8 illustrate the evaluation of grain size values obtained from GEP for the alloys with increasing Cu and Zn concentrations in film and electrolyte relevantly each other, respectively. It is so clear that the formulation obtained from GEP gives well-matched values with literature.

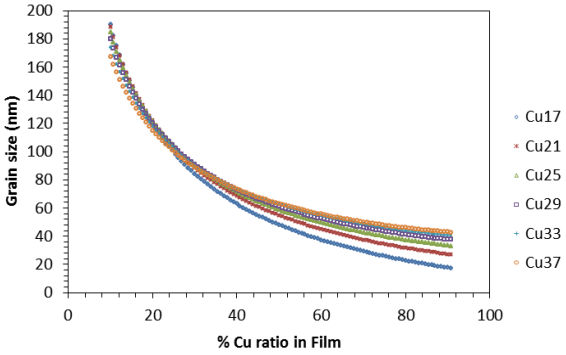

Fig.7 Effect of Cu content both electrolyte and the film on the grain size of electrodeposited Cu-Zn film predicted by GEP for the d-spacing of 2.09 Å.

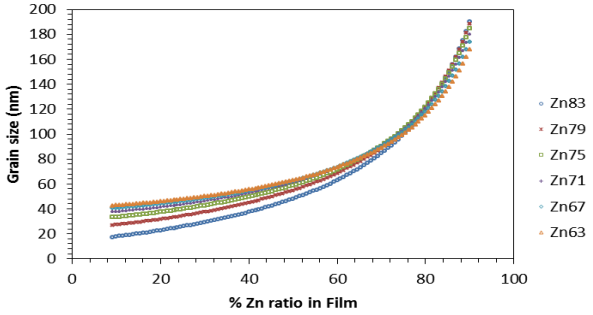

Fig.8 Effect of Zn content both electrolyte and the film on the grain size of electrodeposited Cu-Zn film predicted by GEP for the d-spacing of 2.09 Å.

Fig. 9 demonstrates the dependence of grain size on Cu % ratio in films and d-spacing of the samples obtained from XRD measurements by using GEP formulation. One can say that the grain size by decreases with increasing Cu ratio and increasing d-spacing. Apparently, concentration in the bath and temperature rate significantly affects the formation of the film structure and grain size.

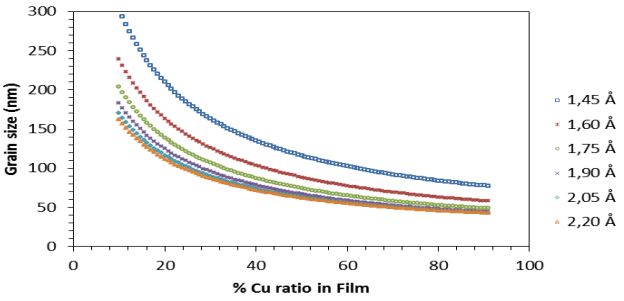

Fig.9 Effect of Cu content in the film and changing d-spacing on the grain size of electrodeposited Cu-Zn film predicted by GEP for the changing FWHM.

6. Conclusion

This present study reports a new and influential approach for the formulations of grain size of the Cu-Zn alloy films using GEP for the first time in the literature. Forty-eight different formulations are proposed for predicting the grain size. The proposed GEP-based formulations are empirical and based on experimental data gathered from our own experiments that not published yet. To predict the grain size of electrodeposited Cu-Zn alloy films, 22. study as given Table 7 is sufficiently accurate. The proposed GEP-based equations are so user friendly that they can be utilized by anybody not absolutely familiar with GEP. The statistical parameters of R^2, MAPE, and RMS show that the proposed GEP-based formulations results has best accuracy and can predict grain size very close to experiment results. The results of this present study will give some useful information to material. GEP may serve as a robust approach and they may open a new era for the accurate and effective explicit formulation of many materials science problems.

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