

OPTIMIZING CHEMISTRY OF BULK METALLIC GLASSES FOR IMPROVED THERMAL STABILITY

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Abstract

Thermo-mechanical-physical properties of bulk metallic glasses (BMGs) depend strongly on the concentrations of each of the chemical elements in a given alloy. The proposed methodology for simultaneously maximizing these multiple properties by accurately determining proper concentrations of each of the alloying elements is based on the use of computational algorithms rather than on traditional experimentation, expert experience, and intuition. Specifically, the proposed BMG design method combines an advanced stochastic multi-objective evolutionary optimization algorithm based on self-adapting response surface methodology and an existing data base of experimentally evaluated BMG properties. During the iterative computational design procedure, a relatively small number of new BMGs need to be manufactured and experimentally evaluated for their properties in order to continuously verify the accuracy of the entire design methodology. Concentrations of most important alloying elements can be predicted so that new BMGs have the best multiple properties. This design concept was verified for superalloys using strictly experimental data. The number of required experimental evaluations of the candidate BMGs with this optimization strategy is expected to be less than 150.

Introduction

Metallic glass is basically an alloy whose metallic species are “frozen” in amorphous glassy state rather than forming a standard crystalline structure. They are several times stronger than steel and considerably harder and more elastic [1]. The bulk metallic glasses (BMGs) are composed of two or more metals in the alloy melt and typically a few diatomatous earth ingredients in order to lower the required cooling rate in order to result in an amorphous structure [2].

Multiple thermo-mechanical properties and cooling speeds of BMG alloys depend strongly on the concentrations of each of the alloying elements. We propose here a novel methodology for predicting the concentration of each of the important alloying elements in BMGs so that the new alloys will have improved thermal stability and glass forming ability. Specifically, we are currently concentrating on simultaneously maximizing liquidus temperature, T_{liq} , and the phase transformation temperature, T_g [2]. The proposed method is based on combining experimentally obtained multiple properties of the BMGs and computational optimization algorithms rather than on traditional experimentation alone, expert experience, and intuition.

Specifically, the proposed BMG design method combines an advanced stochastic multi-objective evolutionary optimization algorithm based on self-organizing graph theory and a self-adapting response surface methodology [3]. During the iterative computational design procedure, a small set of new BMG alloys is periodically predicted, manufactured, and experimentally evaluated for their properties in order to continuously verify the accuracy of the entire design methodology [4-8]. This BMG alloy design optimization method is thus experimentally verified and minimizes the need for costly and time-consuming experimental evaluations of new BMG alloys.

An alternative would be a brute-force optimization of thermo-mechanical properties of BMGs by varying chemical concentrations of N alloying. This approach would involve creating an N-dimensional matrix of alloy compositions and then interpolating and searching for the extreme points in such a matrix. If concentration of each alloying element is varied within a specified range, this variation could be approximated by, for instance, ten parameters. This means that in the case of an alloy with six alloying elements, this “optimization” would require determining properties of $10 \times 6 = 1,000,000$ BMGs each having a different chemical composition. This is obviously impossible and should be replaced by a more economical mathematical optimization.

In order to reduce the number of experimentally evaluated alloys significantly, there has been a strong effort to develop and use several very complex mathematical models that are based on non-equilibrium thermodynamics of solids, thus minimizing the need for manufacturing and experimental evaluation of the actual alloy samples. However, the exclusive use of this strictly computational approach based on artificial neural networks (ANNs) [9] and more recently on genetic algorithms [10] has been shown to possess dubious reliability and versatility as recently frankly demonstrated by Bhadeshia and Sourmail [11]. Furthermore, ANNs are efficient and relatively accurate interpolating algorithms for any multi-parameter function, but they are not efficient and accurate search algorithms and they are definitely not extrapolation algorithms. That is, the use of ANNs alone is not reliable for “getting out of the box” search outside the given data set. Therefore, it is important to understand a need for mathematically sound multi-objective optimization algorithms [12] capable of finding the global minimum and confidently search outside a given initial data base.

Multi-Objective Optimization Algorithm – IOSO

Our approach is based on the use and special adaptation of a stochastic, multi-objective constrained Indirect Optimization based upon Self-Organization (IOSO) algorithm [3]. IOSO multi-objective optimization algorithm allows for concentrations of a number of alloying elements to be optimized so that a finite number of properties of the BMGs are simultaneously extremized [3-8]. The main benefits of this optimizer are its outstanding reliability in avoiding local minimums, its computational speed, and a remarkably small number of required experimental evaluations of BMG samples as compared to more traditional semi-stochastic optimizers such as genetic algorithms. IOSO uses a multi-dimensional response surface technique with adaptive global and middle-range multi-point approximations. The self-adapting response surface formulation used by IOSO allows for incorporation of realistic non-smooth variations of experimentally obtained data and allows for accurate interpolation of such data [4, 8]. The first stage of IOSO is the creation of approximations of the objective functions. Each iteration in this stage represents a decomposition of the initial approximation function into a set of simple approximation functions so that the final response function is a multi-level graph [7]. The second stage of IOSO algorithm is the optimization of these approximation functions. This approach allows for corrective updates of the structure and the parameters of the response surface approximation. This step is followed by manufacturing and experimentally evaluating the thermo-mechanical properties for each of the predicted optimized BMG.

Proposed Design Optimization Methodology and Previous Achievements

First, a finite number of simultaneous objectives should be defined. For example, maximize T_{liq} , maximize T_g , maximize $T_{rg} = T_g / T_{liq}$, minimize cooling speed, minimize overall cost of the raw materials in the BMG, minimize concentrations of most rare and unavailable alloying elements, etc. Second, the minimum and the maximum expected concentrations of a finite number of the most important alloying elements should be specified. If the number of such elements is approximately five or six and the number of simultaneous objectives is two or three, from our experience with optimizing Ni-base superalloys [5-8] we suggest that an initial data base of approximately 100 BMGs has to be developed by utilizing Sobol's algorithm [13] to prescribe chemical concentrations of these alloys. This needs to be done in order to distribute these concentrations in the best possible way so that the consequent multi-dimensional response surface fitting will be maximally accurate with the minimum number of available experimentally evaluated BMGs. These 100 initial BMGs then must be manufactured by casting them in an identical manner. These casts should be then experimentally tested for the specified number of simultaneous objectives. This information is then used for building approximation functions (multi-dimensional response surfaces) which will further be enriched by the IOSO optimizer using modified radial basis functions and multiple artificial neural networks. These approximation functions are then optimized using a non-gradient-based robust multi-objective optimization algorithm IOSO [3]. At each optimization iteration, a multi-criterion optimization task with a specified number of Pareto [12] optimal points (say, 15) needs to be solved. The results of this complex numerical optimization process will be chemical concentrations of, say, 5-6 specified alloying elements in these 15 new BMGs which the optimization algorithm predicted as belonging to the non-dominated Pareto optimal set.

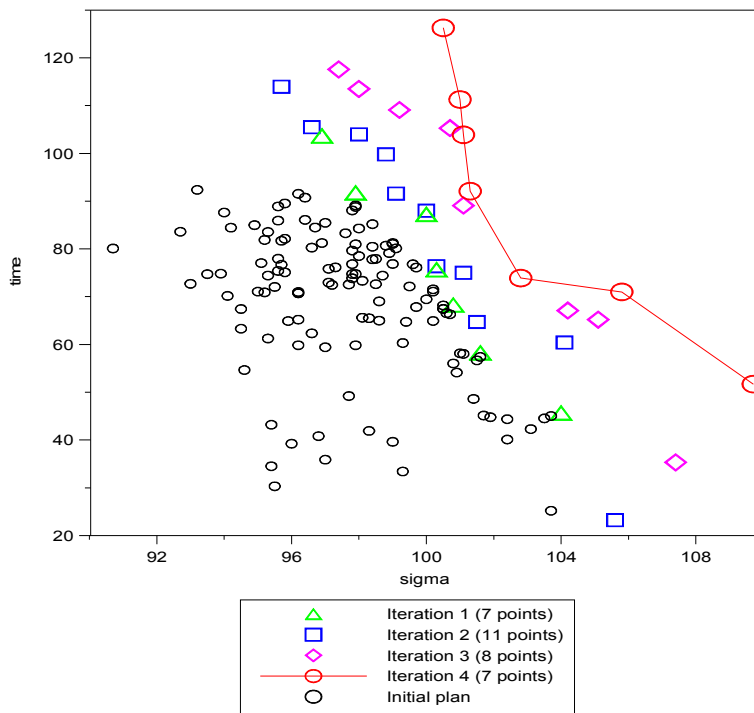


Figure 1. Experimental confirmation of the maximum stress at 20 C and time-to-rupture at 975 C for original 120 Ni-base steel superalloys (small circles) and four generations of 20 optimized Ni-base steel superalloys (other symbols) [7,8].

Since the multi-dimensional response surfaces are fitted using a large number of points created by the artificial neural networks and the radial basis functions, instead of exclusively experimental data, the accuracy of the fit of the response surface will be relatively low.

Consequently, it could be expected that not all of the 15 new optimized BMGs are actually superior to all of the initial 100 BMGs. To clarify this point, these 15 optimized BMGs then need to be manufactured and experimentally evaluated for the multiple properties. This concludes the first design iteration. The second iteration starts by using all (100 + 15 = 115) experimentally evaluated BMGs. The response surface building, enrichment, and optimization process is then repeated using these 115 data points with the same multiple objectives. The 15 newly optimized BMGs then need to be manufactured and experimentally tested to confirm that most of them are better than any of the 115 BMGs used in the second iteration of the design optimization process. The third iteration then starts with all accumulated experimentally tested BMGs (100 + 15 + 15 = 130), repeats all optimization steps and results in 15 new optimized BMGs, and the entire iterative process continues. Figure 1 demonstrates the feasibility of this methodology [1-5].

Because of the unavailability of a large experimental data set for BMGs manufactured in a consistent manner, for the purpose of this study we have decided to create such an experimental data set by combining data from several tables in the works of Prof. Y. Li [14,15] (Table I).

Table I. Experimental Data for BMGs Collected from Works of Prof. Y. Li [14, 15]

#	Zr %	Cu %	Al %	La %	(Cu,Ni) %	Pd %	Si %	Tg(K)	Tl(K)	Trg
1	50	36	14	0	0	0	0	724	1188	0.609
2	50	38	12	0	0	0	0	722	1170	0.617
3	50	40	10	0	0	0	0	714	1176	0.607
4	50	43	7	0	0	0	0	703	1181	0.595
5	46	44	10	0	0	0	0	726	1223	0.594
6	47	45	8	0	0	0	0	714	1218	0.586
7	49	44	7	0	0	0	0	704	1184	0.595
8	48	45	7	0	0	0	0	708	1186	0.597
9	49	45	6	0	0	0	0	704	1187	0.593
10	48	46	6	0	0	0	0	706	1192	0.592
11	49	46	5	0	0	0	0	701	1195	0.587
12	49	47	4	0	0	0	0	697	1208	0.577
13	45	49	6	0	0	0	0	717	1178	0.609
14	45	50	5	0	0	0	0	714	1185	0.603
15	44	51	5	0	0	0	0	719	1189	0.605
16	45	48	7	0	0	0	0	720	1188	0.606
17	45	47	8	0	0	0	0	722	1195	0.604
18	46	49	5	0	0	0	0	711	1193	0.596
19	47	49	4	0	0	0	0	704	1204	0.585
20	54	38	8	0	0	0	0	692	1190	0.581
21	56	36	8	0	0	0	0	685	1212	0.565
22	52	38	10	0	0	0	0	705	1163	0.606
23	54	36	10	0	0	0	0	698	1176	0.594
24	54	40	6	0	0	0	0	684	1216	0.56
25	58	36	6	0	0	0	0	671	1245	0.539
26	0	0	12.4	70	17.6	0	0	403	759	0.53
27	0	0	13.2	68	18.8	0	0	407	742	0.55
28	0	0	14	66	20	0	0	405	674	0.6
29	0	0	14.6	64.6	20.8	0	0	414	696	0.59
30	0	0	15.2	63.1	21.7	0	0	420	699	0.6

31	0	0	15.7	62	22.3	0	0	422	722	0.58
32	0	0	15.9	61.4	22.7	0	0	426	729	0.58
33	0	0	16.3	60.5	23.2	0	0	423	727	0.58
34	0	0	16.6	59.6	23.8	0	0	426	743	0.57
35	0	0	17	58.6	24.4	0	0	431	764	0.56
36	0	0	17.5	57.6	24.9	0	0	435	783	0.56
37	0	0	17.9	56.5	25.6	0	0	440	813	0.54
38	0	0	18.4	55.4	26.2	0	0	436	844	0.52
39	0	0	20.5	50.2	29.3	0	0	435	930	0.47
40	0	0	14	70	16	0	0	404	763	0.53
41	0	0	14	68	18	0	0	405	724	0.56
42	0	0	14	66	20	0	0	405	674	0.6
43	0	0	14	64	22	0	0	411	715	0.57
44	0	0	14	62	24	0	0	417	738	0.57
45	0	0	14	59	27	0	0	422	773	0.55
46	0	0	14	57	29	0	0	427	815	0.52

We then specified that concentrations of all seven alloying elements (Zr, Cu, Al, La, (Cu,Ni), Pd, Si) should be optimized in each of the 46 new Pareto optimal BMGs to be created by the IOSO algorithm while simultaneously maximizing T_g and T_{liq} . Results of this first global iteration with IOSO are the concentrations and T_g and T_{liq} shown in Table II and Figures 2 and 3.

Table II. Concentrations of Alloying Elements, T_g and T_{liq} for 50 Pareto-Optimal BMGs Predicted by IOSO After the First Iteration Using Experimental Data from Table I [14,15].

#	Zr %	Cu %	Al %	La %	(Cu,Ni) %	Pd %	Si %	T_g (K)	T_l (K)	T_{rg}
1	58.000	0.000	0.000	30.189	4.452	0.027	2.291	671.7	1243.3	0.540
2	56.996	9.919	0.000	24.719	3.769	0.002	2.111	672.0	1242.7	0.541
3	58.000	14.664	0.003	17.215	4.617	0.000	1.807	672.1	1242.4	0.541
4	57.820	11.863	0.997	25.583	4.227	0.000	1.636	672.4	1241.8	0.541
5	57.744	12.840	1.280	26.893	4.362	0.000	1.483	672.6	1241.3	0.542
6	57.631	14.539	1.491	28.886	4.161	0.001	1.439	672.8	1240.7	0.542
7	58.000	21.764	0.604	9.096	4.360	0.231	3.211	673.1	1240.2	0.543
8	57.654	15.215	2.087	27.989	4.172	0.000	1.343	673.2	1239.7	0.543
9	56.681	16.364	2.144	31.139	4.032	0.055	1.606	673.5	1239.0	0.543
10	56.710	15.044	2.553	33.863	4.059	0.048	1.699	673.8	1238.4	0.544
11	57.710	26.463	0.090	23.266	0.194	0.176	0.635	674.3	1238.0	0.544
12	57.467	27.146	0.105	24.274	0.158	0.306	0.553	674.7	1237.2	0.545
13	57.359	28.139	0.029	25.283	0.327	0.187	0.790	675.3	1236.1	0.546
14	56.602	28.001	0.121	21.653	0.204	0.564	0.407	675.6	1235.5	0.547
15	56.255	28.317	0.125	19.583	0.203	0.503	0.240	676.0	1234.8	0.547
16	56.124	28.685	0.185	20.111	0.232	0.873	0.394	676.3	1234.1	0.548
17	56.197	29.121	0.169	20.089	0.290	0.963	0.382	676.7	1233.6	0.548
18	56.496	29.745	0.343	16.291	0.000	1.782	0.056	677.0	1232.9	0.549
19	56.230	29.941	0.134	18.502	0.269	1.021	0.231	677.4	1232.3	0.550
20	56.276	30.303	0.412	16.197	0.022	2.037	0.058	677.7	1231.7	0.550
21	56.437	30.951	0.166	16.413	0.233	1.356	0.025	678.3	1230.8	0.551
22	55.982	30.864	0.129	17.015	0.186	1.691	0.015	678.7	1230.3	0.551
23	55.528	30.871	0.216	16.849	0.300	1.590	0.003	679.1	1229.5	0.552
24	55.562	31.317	0.018	17.305	0.065	1.551	0.034	679.7	1228.6	0.553
25	55.482	31.457	0.042	17.544	0.064	1.707	0.031	680.0	1228.1	0.553
26	55.555	31.786	0.012	16.869	0.096	1.396	0.025	680.4	1227.5	0.554

27	55.633	32.308	0.000	16.591	0.398	1.538	0.000	681.2	1226.4	0.555
28	55.648	32.560	0.000	16.397	0.108	1.256	0.001	681.6	1225.8	0.556
29	55.273	32.525	0.012	16.159	0.217	1.244	0.002	682.0	1225.1	0.556
30	54.966	32.587	0.057	16.720	0.213	1.205	0.001	682.4	1224.4	0.557
31	55.166	32.965	0.029	16.576	0.208	1.217	0.000	682.9	1223.7	0.558
32	54.731	32.862	0.000	16.310	0.303	2.025	0.000	683.3	1223.1	0.558
33	54.344	32.808	0.012	18.397	1.031	1.257	0.000	683.7	1222.5	0.559
34	54.190	32.856	0.028	18.817	1.122	1.414	0.001	684.0	1222.0	0.560
35	54.752	33.555	0.134	19.251	1.300	1.279	0.000	684.5	1221.3	0.560
36	54.339	33.436	0.091	17.609	0.255	1.052	0.009	684.9	1220.7	0.561
37	54.618	33.822	0.066	16.965	0.230	0.993	0.000	685.3	1220.2	0.561
38	54.649	34.086	0.079	16.837	0.429	1.508	0.021	685.8	1219.5	0.562
39	54.653	34.423	0.228	13.737	0.260	0.716	0.094	686.3	1218.6	0.563
40	54.596	34.514	0.218	13.335	0.221	0.651	0.092	686.6	1218.2	0.563
41	53.967	34.233	0.168	17.456	0.735	0.785	0.000	687.0	1217.7	0.564
42	54.248	34.538	0.000	20.010	0.000	1.185	0.000	687.4	1217.2	0.565
43	55.477	35.689	0.041	18.214	0.251	1.785	0.011	688.0	1216.6	0.565
44	54.000	34.788	0.088	12.031	0.003	0.540	0.011	688.2	1216.1	0.566
45	54.114	35.039	0.002	12.841	0.013	0.390	0.011	688.7	1215.5	0.566
46	13.810	51.000	16.473	0.000	14.741	60.837	0.007	724.8	1215.5	0.596
47	15.242	50.803	16.863	0.018	15.434	52.467	0.001	724.9	1215.0	0.596
48	15.651	50.975	17.157	0.001	15.115	53.317	0.000	725.0	1214.5	0.597
49	15.494	50.989	17.326	0.180	15.153	55.427	0.000	725.0	1214.2	0.597
50	15.727	51.000	17.515	0.000	15.345	55.044	0.000	725.0	1213.7	0.597

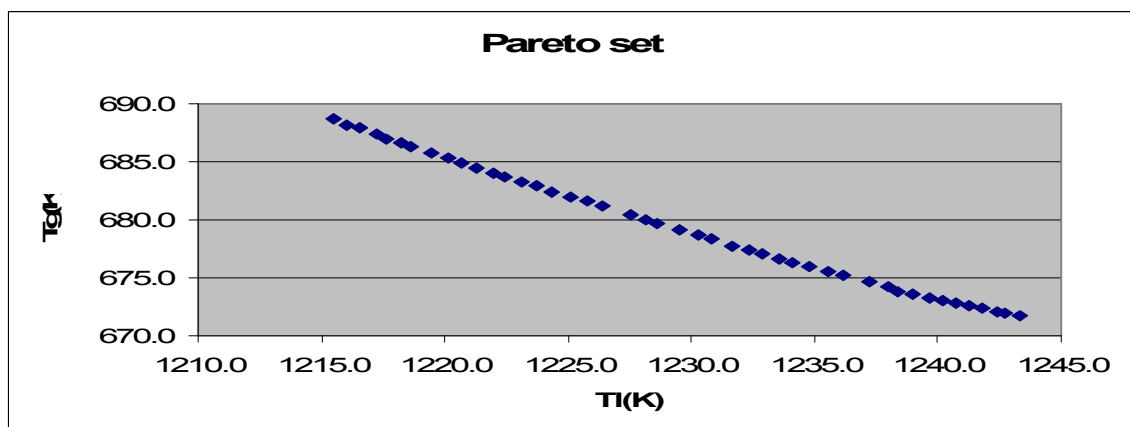


Figure 2. Pareto optimized BMGs when simultaneously maximizing T_g and T_{liq} .

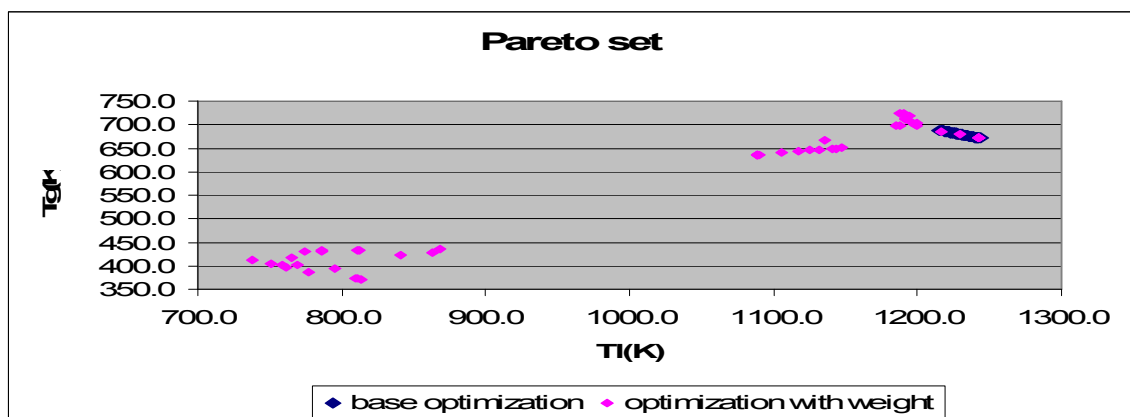


Figure 3. Pareto optimized BMGs when simultaneously maximizing T_g and T_{liq} and minimizing concentrations of each element (Zr, Cu, Al, La, (Cu,Ni), Pd, Si) in each new BMG.

Inverse Design of BMGs for Specified Performance

Conversely, an inverse design option [16] of this computational methodology can offer a capability to design a number of BMG alloys with the same multiple properties, but having different chemistries which will make their availability, cost and utility more affordable. Specifically, we utilized the original experimental data set (Table I) and IOSO optimization algorithm to determine chemical concentrations of Zr, Cu, Al, La, (Cu,Ni), Pd, Si for a number of new BMGs that will all have $T_g = 680$ K and variable T_{liq} (1000 K, 1100 K, 1200 K, 1240 K). Results of such inverse design optimization of BMGs are depicted in Figures 4-11.

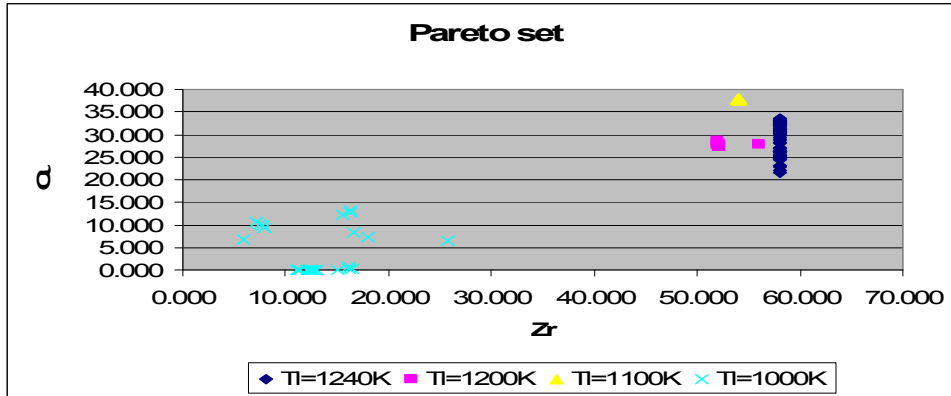


Figure 4. Results of an inverse design optimization problem: $T_g = 680$ K, $T_l =$ variable.

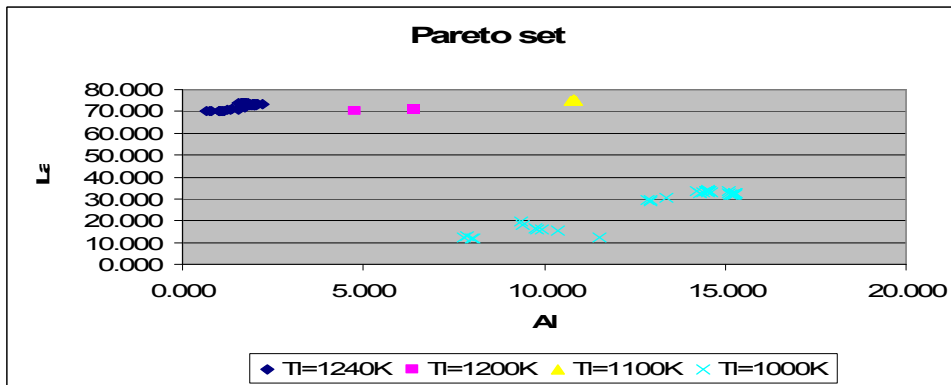


Figure 5. Results of an inverse design optimization problem: $T_g = 680$ K, $T_l =$ variable.

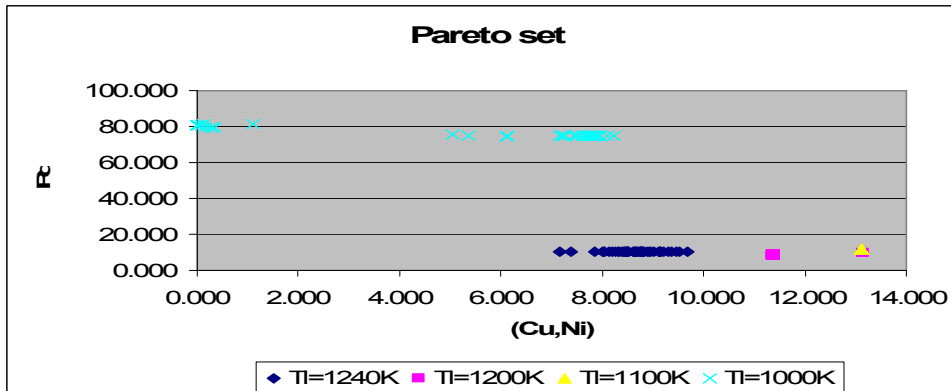


Figure 6. Results of an inverse design optimization problem: $T_g = 680$ K, $T_l =$ variable.

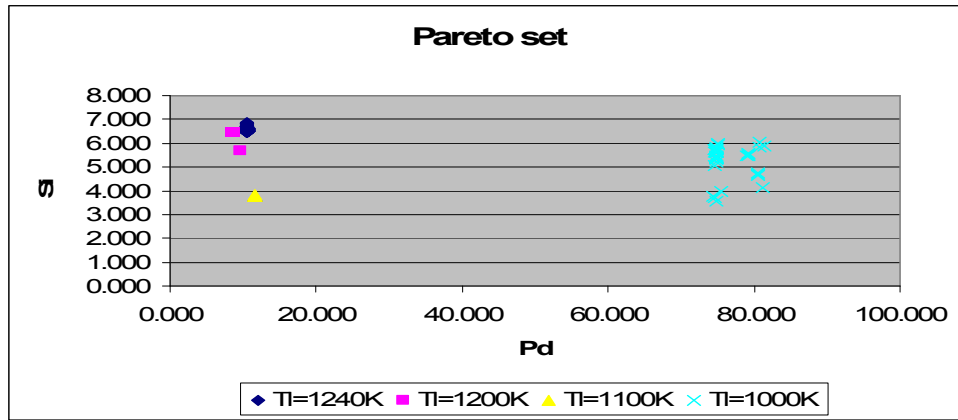


Figure 7. Results of an inverse design optimization problem: $T_g = 680$ K, $T_I =$ variable.

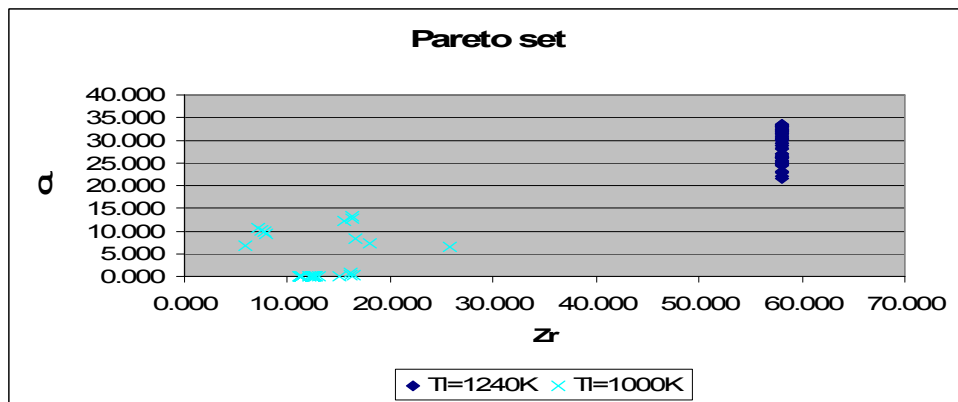


Figure 8. Results of an inverse design optimization problem: $T_g = 680$ K, $T_I =$ variable.

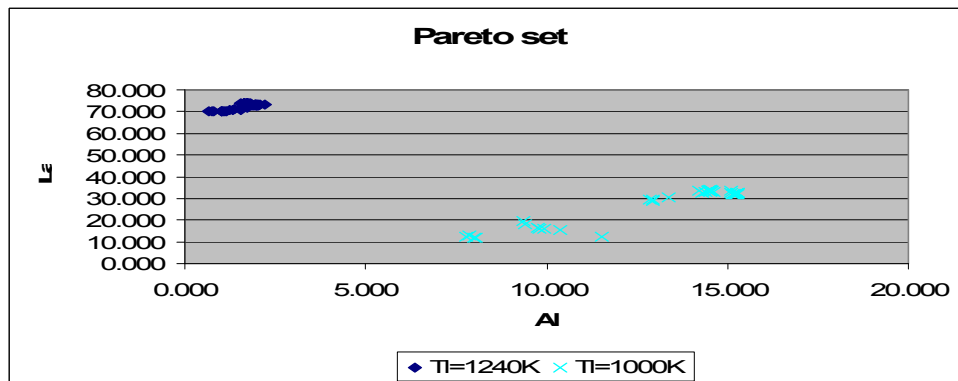


Figure 9. Results of an inverse design optimization problem: $T_g = 680$ K, $T_I =$ variable.

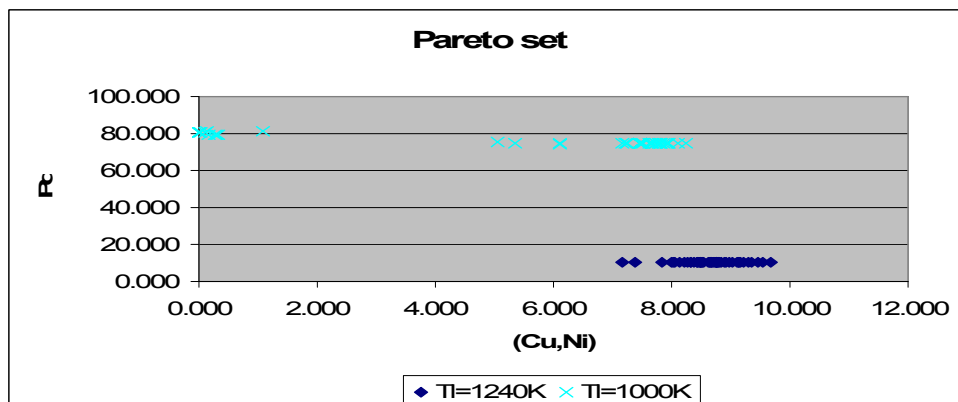


Figure 10. Results of an inverse design optimization problem: $T_g = 680$ K, $T_I =$ variable.

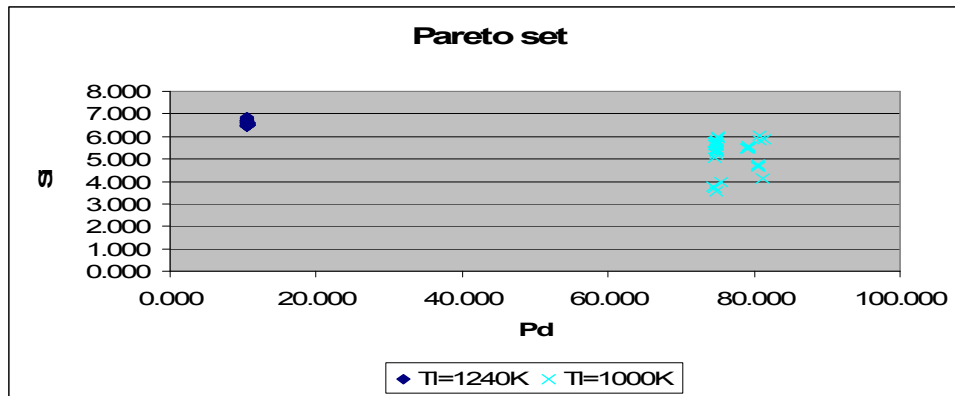


Figure 11. Results of an inverse design optimization problem: $T_g = 680$ K, $T_l =$ variable.

Summary

A new method was demonstrated that offers a realistic possibility to predict chemical concentrations of a number of new BMG alloys so that the new alloys will have superior properties. The new BMG design concept uses a combination of a multi-objective stochastic optimization algorithm and experimental data for thermo-mechanical properties while requiring a minimum number of experimental evaluations of the candidate BMGs, in order to verify the computational results. Conceptually, this design approach could include additional objectives such as minimized cooling speed, maximized T_{rg} , maximized elasticity, minimized cost, etc.

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