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CHEMICAL COMPOSITION DESIGN OF SUPERALLOYS FOR MAXIMUM STRESS, TEMPERATURE, AND TIME-TO-RUPTURE USING SELF-ADAPTING RESPONSE SURFACE OPTIMIZATION

Igor N. Egorov-Yegorov
IOSO Technology Center, Moscow, Russia

George S. Dulikravich
Department of Mechanical and Materials Engineering, Florida International University, Miami, Florida, USA

We have adapted an advanced semistochastic evolutionary algorithm for constrained multiobjective optimization and combined it with experimental testing and verification to determine optimum concentrations of alloying elements in heat-resistant austenitic stainless steel alloys and superalloys that will simultaneously maximize a number of the alloy's mechanical properties. The optimization algorithm allows for a finite number of ingredients in the alloy to be optimized so that a finite number of physical properties of the alloy are either minimized or maximized, while satisfying a finite number of equality and inequality constraints. Alternatively, an inverse design method was developed, which uses the same optimization algorithm to determine chemical compositions of alloys that will be able to sustain a specified level of stress at a given temperature for a specified length of time. The main benefits of the self-adapting response surface optimization algorithm are its outstanding reliability in avoiding local minimums, its computational speed, ability to work with realistic nonsmooth variations of experimentally obtained data and for accurate interpolation of such data, and a significantly reduced number of required experimentally evaluated alloy samples compared with more traditional gradient-based and genetic optimization algorithms. Experimentally preparing samples of the optimized alloys and testing them have verified the superior performance of alloy compositions determined by this multiobjective optimization.

Key Words: Alloy design; Evolutionary optimization; Inverse problems; Material design; Multiobjective optimization; Superalloys.

1. BACKGROUND

Design of superalloys to be used in hot sections of jet engines traditionally involves a lengthy and costly experimental trial-and-error procedure that can easily consume up to 10 years before the new alloy is actually implemented. However, development time for a new aircraft is typically 5 years and has recently been

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Address correspondence to George S. Dulikravich, Department of Mechanical and Materials Engineering, MAIDROC Laboratory, EC 3474, Florida International University, Miami, FL 33027, USA; Fax: +1 (305) 348-6007; E-mail: dulikrav@fiu.edu

reduced to only 4 years. Consequently, the jet engine material designers must develop new alloy design methodologies that will be able to create the new alloys in only a few years or less. During the past several decades of research on the γ/γ' nickel base superalloys, new alloys have been developed by casting, thermally processing, and testing many hundreds and sometimes thousands of different alloy concentrations. Eventually, it was often the use of personal experience and intuition that was used to decide on a small group of finalist concentrations for testing and the winning new alloy concentration adopted for actual commercial use. Thus, it is evident that any reliable predictive modeling and mathematical optimization, at any stage of this empirical alloy design procedure, should be able to reduce the cost and especially the time involved in the development of new alloys with improved performance. The objective of this research was to enable a significant proportion of the development procedure to be done by computation by using the power of mathematical evolutionary optimization techniques in their direct and inverse modes. Although it is understood that the properties of the alloys strongly depend not only on the alloy chemical composition but also on their microstructure, we have decided to leave this important set of influencing parameters for a future more complete alloys design optimization project.

1.1. The Cambridge University Effort

Probably the most prominent center for research activity in certain aspects of predictive modeling and regression analysis in superalloys is at Cambridge University in the United Kingdom [1–8]. Their approach is to use artificial neural network logic for a nonlinear regression analysis where the input data are multiplied by weights, but the sum of all these products forms the argument of a hyperbolic tangent. The output is, therefore, a nonlinear function of the input, the function usually chosen being the hyperbolic tangent because of its flexibility because altering the weights can vary the shape of the hyperbolic tangent. Further degrees of nonlinearity can be introduced by combining several of these hyperbolic tangents, so that the neural network method is able to capture almost arbitrarily nonlinear relationships. For example, it is well known that the effect of chromium on the microstructure of steels is quite different at large concentrations than in dilute alloys. Ordinary regression analysis cannot cope with such changes in the form of relationships.

Using artificial neural networks [8], the Cambridge group has successfully addressed solid solution strengthening, tensile properties, fatigue, creep, lattice misfit in the context of nickel-based superalloys, and has applied the method to other materials and processes. A large number of quantitative models have also been produced by the Cambridge group, dealing with the microstructure and mechanical properties of nickel-based superalloys. They have been used already, both in reducing the scale of experimental programs and in identifying regimens where experiments are essential.

1.2. The Artificial Neural Network

Neural networks are methods for the quantitative recognition of patterns in data, without any a priori specification of the nature of the relationship between

the input and output variables. They can model relationships of almost arbitrary complexity. The outcome of neural network training is a set of coefficients (called weights) and determination of the functions that in combination with the weights relate the input to the output. The training process involves a search for the optimum nonlinear relationship between the inputs and the outputs and is computer intensive. However, once the network is trained, estimation of the outputs for any given inputs is very rapid. There are methods, such as that of MacKay [5], which implement a Bayesian framework on the neural network. The error bars then depend on the specific position in input space, reducing the dangers of extrapolation and interpolation. The Cambridge group has found that this method is capable of revealing interesting metallurgical trends.

The yield and ultimate tensile strength of nickel-based superalloys with γ/γ' microstructures has been modeled [1, 3] by using the neural network method, as a function of the Ni, Cr, Co, Mo, W, Ta, Nb, Al, Ti, Fe, Mn, Si, C, B, and Zr concentrations, and of the test temperature. The analysis was based on data selected from the published literature. The trained models were subjected to a variety of metallurgical tests. As expected, the test temperature (in the range 25–1100°C) was found to be the most significant variable influencing the tensile properties, both via the temperature dependence of strengthening mechanisms and due to variations in the γ' fraction with temperature. Because precipitation hardening is a dominant strengthening mechanism, it was encouraging that the network recognized Ti, Al, and Nb to be key factors controlling the strength.

The Cambridge group methodology for tensile properties has already been exploited at Rolls-Royce to reduce the number of variants involved in experimental alloy design programs. The treatment of iron-based superalloys using both neural network and physical modeling is described by Badmos et al. [7]. A lot of the work and data are available from the materials algorithms project website of Cambridge University [8] at http://www.msm.cam.ac.uk/phase-trans/index.html.

Neural network models in many ways mimic human experience and are capable of learning or being trained to recognize the correct science rather than nonsensical trends. A potential difficulty with the use of regression methods is the possibility of overfitting data. For example, it is possible to produce a neural network model for a completely random set of data. To minimize this difficulty, the experimental data can be divided into two sets, a training data set and a test data set. The model is produced by using only the training data. The test data are then used to check that the model behaves itself when presented with previously unseen data [8].

In addition, artificial neural networks, once fully trained, are very efficient and accurate interpolating algorithms for any multiparameter function. However, this does not mean that the neural networks are automatically efficient and accurate search algorithms or extrapolation algorithms for venturing outside of the available database. These, they are not.

Therefore, it is important to understand a need for mathematically sound multiobjective stochastic optimization algorithms that are capable of finding the global minimum and that can confidently search outside a given initial database.

2. MULTIOBJECTIVE OPTIMIZATION: BACKGROUND

There is a clear and urgent industrywide need for improving material property performance for the applications that they are currently used for and to increase their upper use temperature, strength, and corrosion resistance. The proposed alloy design method takes the new approach of using a stochastic optimization algorithm for optimizing alloy properties with a minimum number of experimental evaluations of the candidate alloys. This approach has the potential of identifying new compositions that cannot be identified without carrying out an unacceptably large number of experiments. Furthermore, this approach has the potential for creating and designing alloys for each application, thereby maximizing their use at reduced cost.

The key to the success of the proposed method is the robustness, accuracy, and efficiency of the multiobjective constrained optimization algorithm. There are only a few commercially available general-purpose optimization software packages. Currently, the most popular commercially available general-purpose optimization software in the United States is iSIGHT [9]. However, these software packages predominantly use a variety of standard gradient-based optimization algorithms, which are known to be unreliable because of their tendency to terminate in the nearest feasible minimum instead of finding a global optimum. Moreover, these optimizers can perform only optimization of a weighted linear combination of objective functions. This formulation does not provide a true multiobjective optimization capability (i.e., each individual objective is not fully extremized). Furthermore, these optimizers require an extremely large number of evaluations of objective functions (mechanical and corrosion properties of alloys), which would make the total number of experimental evaluations unacceptably large because currently available algorithms for confidently predicting physical properties from given alloy concentrations are unavailable. The industry is most probably aware of these drawbacks of the commercially available optimization software. Some people are also becoming aware of the neural network approach to alloy design as practiced at Cambridge University and of the applications of genetic algorithms in materials design [10] and of its coupling with a molecular dynamics simulation approach [11]. However, for the most part, they are not aware of the latest developments in the area of stochastic truly multiobjective constrained optimization because these methods have not been commercialized and have not been demonstrated in this field of application.

The growing need for the multidisciplinary and multiobjective approach to design with a large number of design variables resulted in an increased interest in the use of various versions of hybrid [12], semi-stochastic [13], and stochastic [14–23] optimization algorithms. Including more objectives in the optimization process has similar effects, including more constraints, especially if these constraints are incorporated as penalty functions. With the introduction of the Pareto dominance concept, the possible solutions are divided in two subgroups: the dominated and the nondominated. The solutions belonging to the second group are the "efficient" solutions (i.e., the ones for which it is not possible to improve any individual objective without deteriorating the values of at least some of the remaining objectives). Classical gradient-based optimization algorithms are capable, under strict continuity and derivability hypotheses, of finding the optimal value only

in the case of a single objective. For these algorithms, the problem of finding the group of nondominated solutions (the Pareto front) is reduced to several single-objective optimizations where the objective becomes a weighted combination of the objectives called the utility function. Multiobjective optimization algorithms based on a genetic algorithm have been successfully applied in a number of engineering disciplines. However, for a large number of design variables and objective functions that need to be extremized simultaneously, this approach becomes progressively too time-consuming and unreliable for practical applications in industry.

3. MULTIOBJECTIVE OPTIMIZATION OF ALLOY COMPOSITIONS USING IOSO [18]

We decided to take a new approach of using stochastic optimization algorithm for optimizing alloy properties with minimum number of experimental evaluations of the candidate alloys. The proposed method has the potential of identifying new compositions that cannot be identified without carrying out an unacceptably large number of experiments. Furthermore, the approach has the potential for creating and designing alloys for each application, thereby maximizing their use at reduced cost. The proposed method is based on the use and special adaptation of a new stochastic optimization algorithm specifically for the task of optimizing properties of alloys while minimizing the number of experimental evaluations of the candidate alloys. This multiobjective optimization algorithm is of a semistochastic type incorporating certain aspects of a selective search on a continuously updated multidimensional response surface. Both weighted linear combination of several objectives and true multiobjective formulation options creating Pareto fronts are incorporated in the algorithm. The main benefits of this algorithm are its outstanding reliability in avoiding local minimums, its computational speed, and a significantly reduced number of required experimentally evaluated alloy samples compared with more traditional semistochastic optimizers like genetic algorithms. Furthermore, the self-adapting response surface formulation used in this project allows for incorporation of realistic nonsmooth variations of experimentally obtained data and allows for accurate interpolation of such data.

3.1. Response Surface and Self-Organization Concepts

Our approach is based on the widespread application of the response surface technique with the adaptive use of global and middle-range multipoint approximation. One of the advantages of the proposed approach is the possibility of ensuring good approximating capabilities using minimum available information. This possibility is based on self-organization and evolutionary modeling concepts [18]. During the approximation, the approximation function structure is being evolutionarily changed, so that it allows successful approximation of the optimized functions and constraints having sufficiently complicated topology.

With reference to a particular problem of the creation of an alloy with desirable properties, there will inevitably arise a problem of constraints that need to be specified on the objective functions. These constraints are absent in a more general multiobjective optimization statement. Such objective constraints should be

set by the user (expert) and could be allowed to vary during the solution process. For example, a minimum acceptable value for the Young's modulus of elasticity could be specified as an inequality constraint. Or a maximum acceptable percentage for each of the most expensive ingredients in the alloy could be specified as a cost-objective constraint. In addition, the maximum specific weight of an alloy could be specified as an inequality constraint.

The problem of the search for Pareto-optimum solutions set in the multiobjective optimization while varying chemical composition of an alloy would be an unacceptably labor-intensive process. This is because of an extremely large number of alloy compositions that would need to be created and because several of the properties of each of these alloys would have to be evaluated experimentally. Unfortunately, such problems, as a rule, are difficult to formalize at the initial stage, because the user does not know initially what values of some objectives could be reached and how the remaining objectives will vary. The number of experiments that are necessary for true multiobjective optimization problem solution depends not only on the dimensionality of the problem (the number of ingredient species in an alloy) but also depends to a considerable degree on the topologies of the object functions. Because the user has very little if any a priori knowledge of objective function space topology, it is very difficult to predict the number of experiments required in the optimization application proposed here.

3.2. Summary of Indirect Optimization Based on the Self-Organization (IOSO) Algorithm

Every iteration of IOSO consists of two stages. The first stage is the creation of an approximation of the objective function(s) (Fig. 1).

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 multilevel graph. That is, the evolutionary self-organizing algorithms are based on the modified version of the method of accounting for the groups of arguments. Such algorithms use the evolutionary procedure of constructing approximation functions in the form of multilevel graphs (Fig. 2) and solving the structure-parametric approximation problem in the process.

The second stage is the optimization of this approximation function. This approach allows for corrective updates of the structure and the parameters of the response surface approximation. The distinctive feature of this approach is

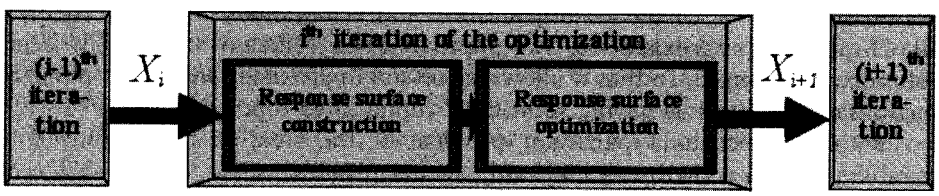

Figure 1 IOSO iteration scheme.

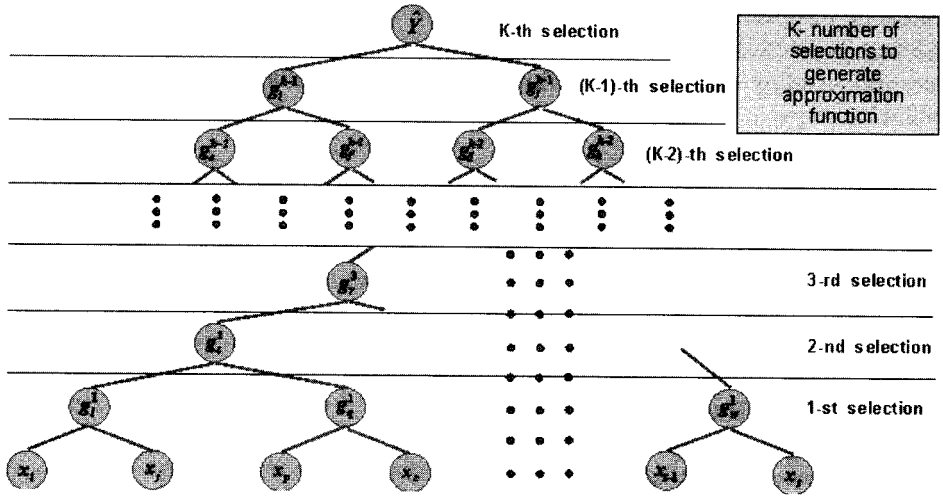

Figure 2 Example of the IOSO response surface structure.

an extremely low number of trial points to initialize the algorithm. During each iteration of IOSO, the optimization of the response function is performed only within the current search area. This step is followed by an actual experimental evaluation for the obtained point. During the IOSO operation, the information concerning the behavior of the objective function in the vicinity of the extremum is stored, and the response function is made more accurate only for this search area. Thus, during each iteration, a series of approximation functions (Fig. 3) for a particular objective is built. These functions differ from each other according to both structure and definition range.

The subsequent optimization of these approximation functions allows us to determine a set of vectors of optimized variables. IOSO using Sobol's algorithm [24] was used for redistribution of the initial points in the multidimensional function space. IOSO also includes algorithms of artificial neural networks (ANN) that use appropriately modified radial-basis functions to enrich the original data set and build the response surfaces. The modifications consisted in the selection of ANN parameters at the stage of their training that are based on the

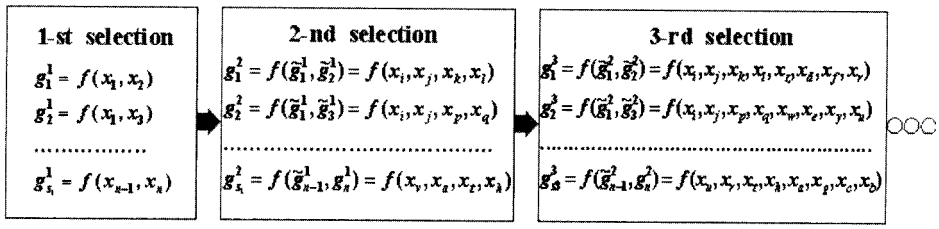

Figure 3 IOSO approximation process scheme.

following two criteria: minimal curvature of response surface and provision of the best predictive properties for a given subset of test points, $W_{best} \in W_{ini}$. Each iteration of alloy composition multiobjective optimization technique involves the following steps.

1. Building and training ANN1 for a given set of test points proceeding from the requirement $W_{best} = W_{ini}$.
2. Conducting multiobjective optimization with the use of ANN1 and obtaining a specified number of Pareto-optimal solutions P_1.
3. Determining a subset of test points W_{best} that are maximally close to points P_1 in the space of variable parameters.
4. Training ANN2 proceeding from the requirement to provide the best predictive properties for obtained subset of test points $W_{best} \in W_{ini}$.
5. Conducting multiobjective optimization with the use of ANN2 and obtaining a set of Pareto-optimal solutions P_2.

In general, the database contains information on experimentally obtained alloy properties compiled from different sources and obtained under different experimental conditions. As a result, for alloys with the same chemical compositions, there can be considerable differences of measured properties. These differences can be explained as errors due to the particular conditions existing during the experiments (measurement errors), and by the effect of certain operating conditions (e.g., thermal condition of alloy making). Unless operating conditions are quantified numerically, their influence is regarded as an additional chance factor. In its simplified form, the methodology can be presented as the following set of actions:

1. Formulation of optimization task (i.e., selection of variable parameters) definition of optimization objectives and constraints, and setting initial (preliminary) ranges of variable parameters variations.
2. Preliminary reduction of the experimental database. At this stage, the points meeting the optimization task statement are picked up from the database so that alloys having chemical composition outside the chosen set of variable parameters are rejected. Alloys for which there are no data for at least one optimization objective are rejected. In addition, alloys with chemical compositions outside the set range of variable parameters are also rejected.
3. Final reduction of the experimental database. Because accuracy of the building of response surfaces substantially depends on uniformity of distribution of variable parameters in the surveyed area, rejection of experimental data points falling outside of the universal set is performed. At the end of this stage, a final range of variable parameters for optimization is set.
4. Execution of multiobjective optimization resulting in a specified number of Pareto-optimal solutions.
5. Analysis of optimization results.
6. Carrying out an experiment to obtain a set of Pareto-optimal alloy compositions (or a certain subset) and analysis of the results obtained.

7. Change of optimization problem statement (number of simultaneous objectives and constraints, the set and range of variable parameters) and returning to step 2.
8. Modification of database and returning to step 4.
9. Stop.

4. EXAMPLES OF DIRECT DESIGN OF ALLOYS USING IOSO OPTIMIZATION

A preliminary proof of this alloy optimization concept was published recently by Dulikravich et al. [25] and expanded more recently to a larger data set by Egorov and Dulikravich [26]. For this particular case, the initial database was procured containing information on 201 experimentally tested alloys. A preliminary analysis of data has shown that for certain alloys there is no complete information on alloy chemical composition. Such alloys were excluded from further analysis. Besides, some chemical elements (V, Bi, Se, Zr, Sb, Cd) were present in a very small number of alloys, which makes it impossible to assess their effect from information in this database. Such alloys were also excluded from further analysis. The remaining database had 176 alloys. At the next stage, an evaluation of uniformity of distribution of the percentage values of different elements in the existing range was made. It turned out that certain alloying elements had concentrations differing very strongly from the universal set. For example, percentage of sulfur in one of the alloys exceeding average value by some 10 times. Such alloys were excluded from further analysis. The remaining database had 158 alloys.

The following parameters were then used as optimization objectives: stress (PSI-maximize); operating temperature (T-maximize); time to "survive" until rupture (Hours-maximize). During this research, the solution of a simultaneous three-objective optimization problem and a series of two-objective problems were accomplished when one of the considered parameters was constrained.

4.1. Influence of the Number of Alloying Elements

In this problem, the percentages of the following 17 alloying elements were taken as independent variables: C, S, P, Cr, Ni, Mn, Si, Cu, Mo, Pb, Co, Cb, W, Sn, Al, Zn, and Ti. The ranges of these elements were set as follows. First, minimum and maximum values for the existing set of experimental data (Exp_min_i, Exp_max_i, $i = \overline{1, 17}$) were defined. Then, new minimum and maximum values for each of the 17 elements were obtained according to the following simple dependencies: ($Min_i = 0.9 \cdot Exp\_min_i$, $Max_i = 1.1 \cdot Exp\_max_i$, $i = \overline{1, 17}$). The allowable ranges are given in Table 1. Although the lower range for Cr and Ni content almost corresponds to AISI 310 scale-resistant stainless steel, the upper range of Cr and Ni content correspond to superalloys [27]. It should be pointed out that the chemistry of the two types of alloys is entirely different.

The three-objectives optimization run was then repeated with only the following nine chemical elements as independent variables: C, Cr, Ni, Mn, Si, Mo, Cb, W, and Ti. We have followed the same steps during the optimization as when solving the problem with 17 variables. But, in this case, there were noticeable differences due to accuracy deterioration of the response surface representation. Thus, when using fewer alloying elements, while decreasing the number of variables for the same experimental data set, additional noise was introduced in this data set.

Table 1 Ranges of variation of 17 independent variables (chemical elements in the steel alloy)

	min	max
C	0.063	0.539
S	0.001	0.014
P	0.009	0.031
Cr	17.500	39.800
Ni	19.300	51.600
Mn	0.585	1.670
Si	0.074	2.150
Al	0.001	0.075
Mo	0.000	0.132
Co	0.000	0.319
Cb	0.000	1.390
W	0.000	0.484
Sn	0.000	0.007
Zn	0.001	0.015
Ti	0.000	0.198
Cu	0.016	0.165
Pb	0.001	0.006

4.2. Simultaneous Optimization of Three Objectives for Alloys Having 17 Chemical Elements

During the first stage, the problem of simultaneously optimizing three objectives was solved with 100 points of Pareto-optimal solutions. Figure 4 presents the obtained Pareto-optimal solutions in objectives' space (PSI-HOURS). Analysis of this figure allows us to extract an area of admissible combinations of different optimization objectives. It can be seen that results are distributed in the admissible part of the objectives' space quite uniformly. Such a distribution offers a possibility

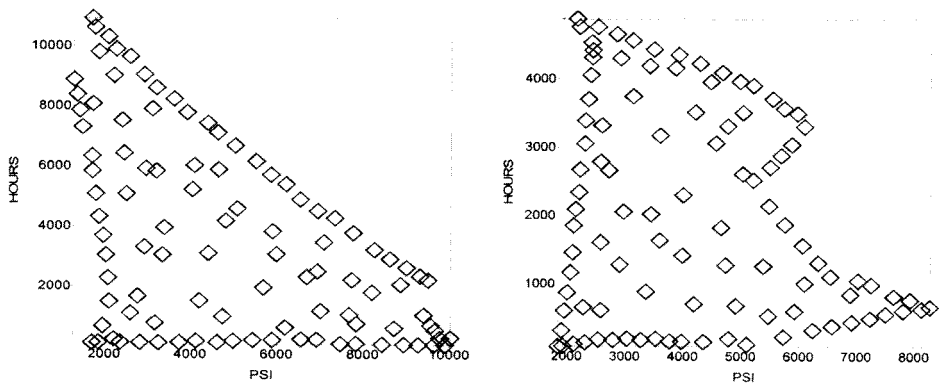

Figure 4 Time to rupture versus strength interdependence of optimization objectives for three-objectives Pareto set with 17 chemical elements (left) and with 9 chemical elements (right).

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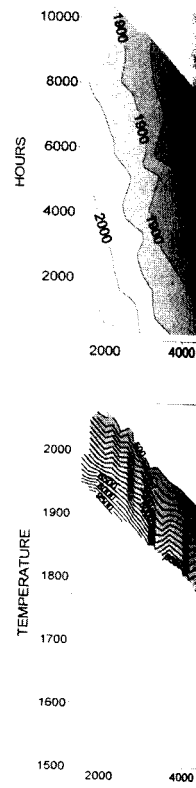

Figure 5 Time-to-rup
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for a significant improvement of accuracy of response surfaces on condition that the experiments will be carried out at the obtained Pareto-optimal points. In principle, the first iteration of the process of alloy chemical composition optimization by several objectives could be regarded as completed. Then, in accordance with the elaborated technique, it is necessary to conduct experiments at the obtained Pareto optimal points, evaluate accuracy of prediction of values of partial optimization criteria, and either complete the process or perform another iteration. However, such a strategy seems very difficult to implement for a researcher who knows his tasks more accurately. It can be seen that the ranges of variation of optimization objectives for the obtained Pareto set are very wide. At the same time, if a researcher can formulate the problem more specifically (e.g., by setting constraints on the objectives) the volume of experimental work can be substantially reduced.

Figure 5 presents interdependence of the chosen optimization objectives built on the obtained set of Pareto-optimal solutions. Figure 6 demonstrates the difference in topology of the multi-objective function space when using different numbers of alloying elements. Larsen-Mueller diagram (Fig. 7) has

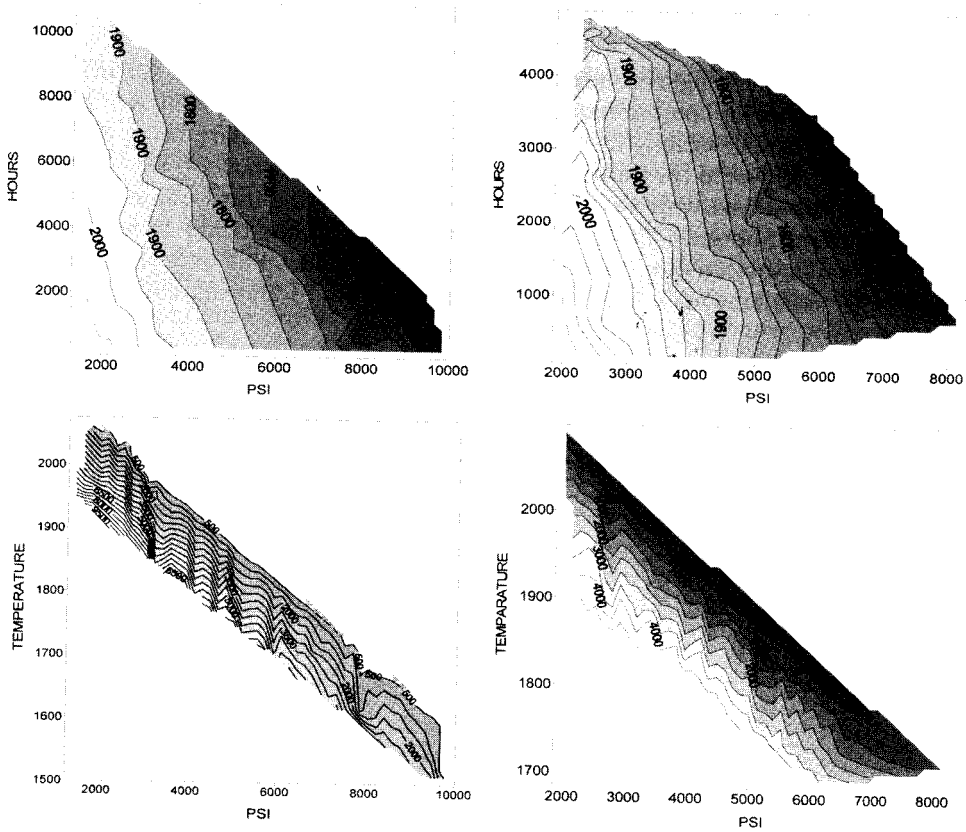

Figure 5 Time-to-rupture versus strength and temperature versus strength interdependences of optimization objectives for Pareto set resulting from a three-objectives optimization with 17 chemical elements (left) and with 9 chemical elements (right).

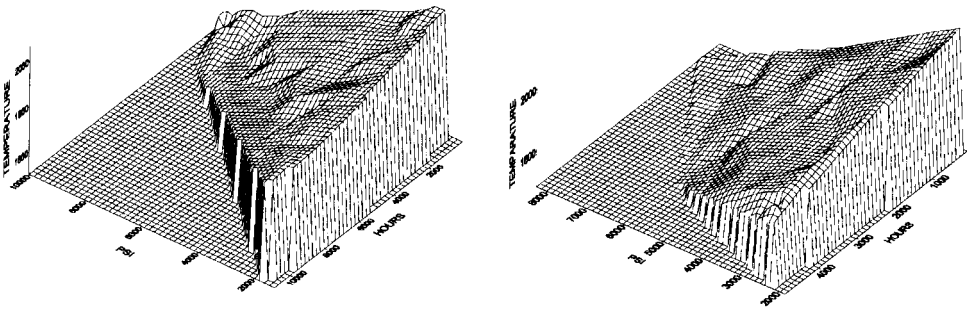

Figure 6 Topography of response surfaces of three-objective optimization problems with 17 chemical elements (left) and with 9 chemical elements (right).

PSI on the vertical axis and the following expression on the horizontal axis (temperature in Rankine degrees) $*$ log(HOURS + 20). Here, logarithm is with the basis 10, whereas temperature is in Rankine = temperature in Fahrenheit + 460. Figure 8 illustrates the general trend in the abilities of the optimizer to create alloys with superior performance as a function of the number of alloying elements chosen for optimization.

Analysis of these figures shows that the increase of temperature, for instance, leads to a decrease of compromise possibilities between PSI and HOURS. Hence, if a researcher knows exactly in what temperature range the alloy being designed will be used, it would be more economical that a sequence of two-objective optimization be solved with an additional constraint for the third objective. Thus, a more efficient approach to optimizing alloy compositions could be to solve a sequence of two-objective optimization problems in which PSI and HOURS are regarded as simultaneous objectives, while imposing the following constraints on temperature:

- Problem 2. $T \geq 780$C (1600 F), number of Pareto-optimal solutions is 20.
- Problem 3. $T \geq 982$C (1800 F), number of Pareto-optimal solutions is 20.

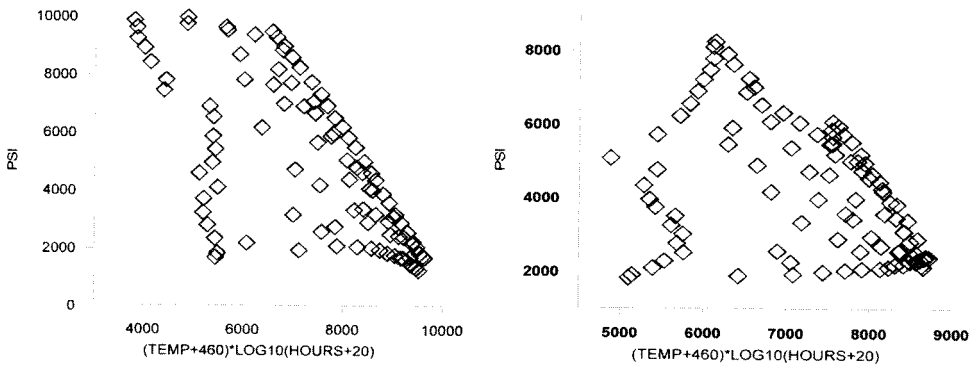

Figure 7 Larsen-Mueller diagram for Pareto sets resulting from a three-objective optimization with 17 chemical elements (left) and with 9 chemical elements (right).