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# CALPHAD: Computer Coupling of Phase Diagrams and Thermochemistry

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## Accelerated exploration of multi-principal element alloys for structural applications

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### ABSTRACT

A strategy for accelerated discovery and exploration of multi-principal element alloys was developed and used to identify new alloys within a design window of desired microstructures and properties. As an example, the strategy was applied to analyze thousands of 3- 4-, 5- and 6-component alloys at equiatomic compositions of the alloying elements. Currently available thermodynamic databases were used to assess equilibrium phase diagrams for these alloys. The validity and reliability of the calculated phase diagrams were estimated based on the extent of experimental binary and ternary data used to build the respective thermodynamic databases. Alloys with specific characteristics, such as single-phase solid solution alloys with the use temperature above 1000 °C, were identified using an automated analysis of the calculated phase diagrams. The density, elastic moduli and costs of these alloys were estimated using the rule of mixtures of pure elements and were used as additional criteria for alloy selection. This approach allowed rapid, albeit preliminary, screening of many thousands of alloys and identification of promising candidate compositions, some of which are reported in this paper, for more time intensive experimental validations and assessments.

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### 1. Introduction

Materials with high strength, low density and acceptable damage tolerance are in high demand for transportation and energy applications. Four alloy families are commonly used to meet the operating requirements in these applications. In order of increasing application temperature, these are aluminum alloys, titanium alloys, iron alloys (steels and superalloys) and nickel-based superalloys. Alloy density generally increases with increasing use temperature. These alloys are each based on a single element with additional elements added to improve the balance of properties. The total concentration of secondary elements generally does not exceed 10–15%, but in superalloys it can be as high as 40% [1].

Ni-based superalloys have been the material of choice for over 70 years in load-bearing applications at the highest temperatures, especially in fracture-critical components. Alloy density has increased with the gradual evolutionary changes in alloy chemistry needed to improve the balance of properties (creep, strength, fatigue) at higher temperatures [2]. Moreover, operating temperatures of Ni-based superalloys are now reaching theoretical limits, controlled by incipient melting at temperatures in the range of 1200–1300 °C. New materials with higher strength, higher operating temperatures and/or lower density are in high demand.

Numerous concepts have been pursued in the past 50 years to surpass the balance of properties offered by these exceptional materials, but none have been successful in displacing superalloys [3].

Recently there has been a great deal of interest in a new alloy design strategy focused on multi-principal-element alloys (MPEA) where 3–6 elements are added at near equal atomic amounts [4–8]. This approach opens up a large composition space that has previously been unexplored and necessitates the use of a rapid screening technique to down select compositions that would give the best balance of properties. This assumes a-priori knowledge of the best combinations of target phases and known metallurgical principles provide some guidance. One variant of this approach, known as High Entropy Alloys (HEAs), includes alloys containing 5 or more alloying elements at or near equiatomic amounts [4–6] and is focused on stabilizing a single solid solution phase. HEAs have interesting and unexpected microstructures and properties [4,5,7,9–11]. In spite of the compositional complexity, many HEAs have the tendency to form solid solution (SS) phases and the number of phases observed in these alloys is much less than the maximum predicted from the Gibbs phase rule [5,9,12–14]. Formation of a disordered SS phase in favor of an ordered IM phase in a HEA is explained [4,5,9–11] by higher entropy of mixing of alloying elements,  $\Delta S_{\text{mix}}$ , in the multicomponent SS phase, relative to the entropy of formation,  $\Delta S_f$ , of a competing IM compound. The difference between  $\Delta S_{\text{mix}}$  and  $\Delta S_f$  increases with an increase

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in the number of the alloying elements and reaches its maximum at equiatomic compositions. Thus the reduction of the Gibbs free energy by the entropy term occurs with an increase in temperature more rapidly in SS than in IM phases. The HEA approach could be a powerful tool for producing new SS-based high-temperature structural alloys in an alloy composition space that has not been previously explored.

A brief survey of the literature shows significant alloy exploration in both face centered cubic (FCC) and body centered cubic (BCC) crystals. In the FCC's several groups have produced and characterized HEAs containing Al, Co, Cr, Fe, Mn, Ni and Cu [14]. These can be considered an extension of austenitic stainless steels or superalloys, which have Fe, Cr and Ni as their major constituents but may also contain significant concentrations of other elements. Another group of HEAs reported in the literature is refractory containing HEAs [15,16]. The major phases in these alloys have body-centered cubic (BCC) crystal structures and minor phases are mainly topologically close packed Laves phases. The BCC-based HEAs are much stronger than FCC-based HEAs, but many of them experience brittle-to-ductile transition having low ductility at room temperature, especially in the as-cast condition.

As the entropy term of the Gibbs free energy decreases linearly with a decrease in temperature, high-temperature SS phases are expected to become metastable at lower temperatures. Indeed, detailed microstructure analysis revealed formation of multiphase structures consisting of SS and IM phases in many HEAs after annealing and/or thermo-mechanical treatment at temperatures below 1000–1200 °C, and only few HEAs retained SS structures [17–21]. Unfortunately, experimental studies of equilibrium phases in HEAs at  $T < 1000$  °C are challenged by slow diffusion kinetics [22]. At the same time, only alloys with no first order phase transformations in the temperature range of the application use should be selected to ensure structural stability. Therefore, equilibrium phase diagrams of these new alloys must be available.

In spite the large number, over  $\sim 200$ , of new MPEAs reported (i.e., in average,  $\sim 20$ – $25$  alloys per year), a vast number of compositional possibilities have yet to be explored. Indeed, the amount of alloy systems increases with factorial speed when the number of principal alloying elements increases. For example, a pallet of 27 elements results in 27 alloy systems with a single principal element, 2925 alloy systems with 3 principal elements and 296,010 alloy systems with 6 principal elements. It is impossible to evaluate such huge number of alloy systems within a reasonable period of time using traditional methods. For example, if one alloy can be made and characterized in one week, it would take  $\sim 20$  years to evaluate 1000 alloys, and it is not certain that some of these alloys would have properties superior to currently available commercial alloys. New approaches are required to quickly and efficiently screen promising alloy compositions for particular applications.

A strategy to design and evaluate MPEAs for structural use in the transportation and energy industries has recently been proposed [7]. A systematic design approach uses palettes of elements chosen to meet target properties and gives methods to build MPEAs from these palettes. The developed strategy includes both single-phase SS MPEAs, as well as two phase systems consisting of a SS matrix and an intermetallic phase for precipitate hardening. To systematically screen and evaluate the vast composition space offered by MPEAs and HEAs, high-throughput computations and experiments with a feedback loop for validation have been suggested [7]. The idea is to quickly reject systems with some critical property deficiency and to focus resources on characterizing the remaining systems.

In this work, we use the proposed strategy [7] to develop a methodology to quickly identify, screen and analyze hundred thousand alloy compositions for required microstructure and

properties. To achieve this goal, screening criteria for alloy selection are proposed, CALPHAD calculations are used to identify the critical thermodynamic properties and microstructural features, and computer programs are developed to automate alloy selection, property calculations, analysis and screening processes. Due to a huge number of possible MPEA compositions, only equiatomic MPEAs were analyzed in this work to validate the proposed screening approach. Expanding our approach to the non-equiatomic composition space is our next step, which we are currently pursuing and which is beyond the scope of this paper.

## 2. Approaches and methods

### 2.1. Identification of elements and alloy properties for screening

As the first step, a master list of elements for prospective MPEA systems that can be used in intermediate temperature (MT) and high temperature (HT) applications has been identified [7]. Goal properties for these two MPEA families should exceed the characteristic properties of Ti and Ni alloys, respectively, by a margin sufficient to warrant the extra risk and cost of development, scale-up, certification and insertion. Thus the alloying elements are selected to satisfy the requirements of the target application window based on their intrinsic properties. Starting with the periodic table of elements and excluding all non-metals, halogens, noble gases, toxic and radioactive elements, semi-metals (except Si), elements with the melting temperature,  $T_m$ , less than 900 °C (except low density Mg and Al), as well as extremely rare, costly, dense or compliant elements, 27 elements have been selected. These are Ag, Al, Co, Cr, Cu, Dy, Fe, Gd, Hf, Lu, Mg, Mn, Mo, Nb, Ni, Re, Rh, Ru, Sc, Si, Ta, Ti, Tm, V, W, Y, and Zr. The reasons why low-density Mg and Al are left in the list are discussed in our previous paper [7]. To develop the methodology for alloy selection and to validate the approach, only equiatomic-composition alloys (hereafter we call them *equiatomic alloys*) with 3–6 elements from the selected list have been evaluated in this work. Thus the processed MPEA systems include not only HEAs with 5 or more elements, but also 3- and 4-component MPEAs. The number of 3-, 4-, 5-, and 6-component equiatomic alloys that can be produced from the selected 27 elements are 2925, 17,550, 80,730, and 296,010, respectively, i.e. 397,215 alloys.

As the second step, the alloy properties used for alloy selection and the methods to calculate these properties have been identified. The first set of screened properties includes thermodynamic properties of an alloy. For example, only alloys with no first order phase transformations in the temperature range of the application use should be selected to ensure structural stability. Therefore, all phase reaction temperatures, if present, must be above the maximum use temperature,  $T_{use}$ . Good ductility and fracture toughness require a solid-solution (SS) primary phase, while potent strengthening mechanisms (e.g. SS and precipitation strengthening) may require a second (SS or intermetallic, IM) phase that can be dissolved and re-precipitated above  $T_{use}$  but below solidus,  $T_m$ . The number and volume fraction of IM phases should be restricted to retain acceptable ductility and fracture toughness. Thus the temperatures and types of all solid state reactions, as well as the number and type of phases at  $T_m$  and below  $T_{use}$  should be estimated for every selected alloy. These properties cannot be estimated using the rule of mixtures of pure elements and CALPHAD calculations are used as outlined in the next section. For a given alloy,  $T_{use}$  is the lower of 80% of absolute  $T_m$  and the lowest temperature below which no solid state phase reactions occur.

The second set of properties used for the alloy screening includes alloy density,  $\rho$ , elastic modulus,  $E$ , and cost,  $P$ . They are calculated using the rule of mixtures and the respective properties

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