Materials and Design 143 (2018) 185–195

Contents lists available at ScienceDirect

Materials and Design

journal homepage: www.elsevier.com/locate/matdes

Evolutionary design of strong and stable high entropy alloys using multi-objective optimisation based on physical models, statistics and thermodynamics

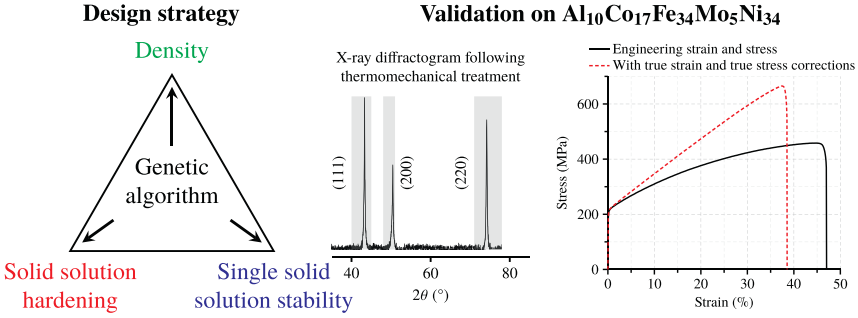
Edern Menou[a,b], Isaac Toda-Caraballo[c,d], Pedro Eduardo Jose Rivera-Díaz-del-Castillo[e], Camille Pineau[a], Emmanuel Bertrand[a], Gérard Ramstein[b], Franck Tancret[a,*]

[a] Institut des Matériaux Jean Rouxel (IMN), Université de Nantes, CNRS, 2 rue de la Houssinière, BP 32229, Nantes Cedex 3 44322, France
[b] Laboratoire des Sciences du Numérique de Nantes (LS2N), Université de Nantes, CNRS, Rue Christian Pauc, BP 50609, Nantes Cedex 3 44306, France
[c] University of Cambridge, Department of Materials Science and Metallurgy, Charles Babbage Road, Cambridge CB3 0FS, United Kingdom
[d] Materalia Research Group, Department of Physical Metallurgy, National Centre for Metallurgical Research (CENIM-CSIC), Gregorio del Amo 8, 28040 Madrid, Spain
[e] Department of Engineering, Lancaster University, Lancaster LA1 4YW, UK

HIGHLIGHTS

- Models for single-phase stability and solid solution strengthening are integrated within a multi-objective genetic algorithm.
- Thousands of non-equimolar face-centered-cubic single-phase alloys are designed using this framework.
- Experiments confirm that the specific strength of one designed alloy surpasses that of alloys with comparable microstructure.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 11 September 2017
Received in revised form 19 January 2018
Accepted 22 January 2018
Available online xxxx

Keywords:
Gaussian process
CALPHAD
AlCoFeMoNi
NSGA-II

ABSTRACT

A new integrated computational HEA design strategy is proposed. It combines a multi-objective genetic algorithm with (i) statistical criteria to guide the formation of a single phase, supplemented by computational thermodynamics (Thermo-Calc) and (ii) models for the estimation of alloy yield stress *via* solid solution hardening, to be maximised, and alloy density, to be minimised. This strategy is applied to the design of face-centered-cubic (FCC) HEAs and yields several thousands of new alloys. An alloy featuring an interesting combination of predicted stability, strength and density, $Al_{10}Co_{17}Fe_{34}Mo_5Ni_{34}$ (at%), is chosen among them, fabricated by vacuum arc melting and experimentally tested. The microstructure of this new HEA consists in a single FCC solid solution, as evidenced by X-ray diffraction (XRD), scanning electron microscopy (SEM) and X-ray energy dispersive spectroscopy (EDS) mapping. With a density of 7.95 $g \cdot cm^{-3}$, a Vickers hardness of 1.78 GPa, a yield stress of 215 MPa and an ultimate tensile strength of 665 MPa in the annealed state, its properties surpass those of existing FCC HEAs of comparable density.

1. Introduction and background

For the past twenty years, interest in highly concentrated multi-component alloys (HCMAs) and multi-principal element alloys forming single solid solutions, also known as "high entropy alloys" [1], has been continuously increasing. As per Yeh's definition, these

* Corresponding author at: Institut des Matériaux Jean Rouxel (IMN), Université de Nantes, CNRS, 2 rue de la Houssinière, BP 32229, Nantes Cedex 3 44322, France.
E-mail addresses: edern.menou@univ-nantes.fr (E. Menou), franck.tancret@univ-nantes.fr (F. Tancret).

https://doi.org/10.1016/j.matdes.2018.01.045
0264-1275/

alloys should contain at least five different elements whose content ranges from 5 to 35 at %; therefore, they exhibit a large configurational entropy which should help stabilising a single solid solution. Although later works showed that the sole configurational entropy is not a major requirement for the stability of a solid solution [2], the designation "HEA" remains preponderant in the literature and will be used in this paper indistinguishably from "single solid solution highly concentrated multicomponent alloys".

Growing interest in high entropy alloys is motivated by the unique combinations of properties they can feature [3,4]: low density, high strength, high ductility, good corrosion resistance, etc. To date, their development has mainly relied on trial-and-error experiments following some physicochemical rules [5–11] and more recently on modelling [12–14], which provided discrete predictions or even ways to systematically explore given alloy systems, sometimes with the help of CALPHAD (CALculation of Phase Diagrams) predictions [15–19]. The desirable feature is generally the formation of a single, stable solid solution. In this respect, a robust and comprehensive method has recently been proposed to predict with a good level of confidence the formation of a stable solid solution in HCMAs [20]. Yet, the design of high performance HEAs must not rely uniquely on phase stability, just as the design of "traditional" alloys generally involves sets of requirements. Mechanical properties in relation to density should also be taken into account with the ideal aim of maximising the former or minimising the latter. In this regard, a recently proposed model enables the estimation of the solid solution hardening (SSH) contribution in such alloys [21], while the density can be computed using a rule of mixtures. Being able to predict phase stability, strength and density is a requirement for the alloy design exposed in this paper, and the corresponding models will be briefly overviewed in the next section.

However, the titanic number of potential HEAs forbids even a computer-aided screening of all possible non-equimolar compositions using the aforementioned models. Indeed, considering around fifty metallic elements of the periodic table can enter the composition of HEAs, an impressive number of alloy families, *i.e.*, sets of different elements, can be enumerated: picking up five elements amongst the fifty leads to more than two million families (C_5^{50}); this already amounts to over two million possible equimolar alloys with five elements. Varying the concentration of each element by small steps between 5 and 35 at% brings the total number of producible alloys to thousands of billions. For instance, about 1.2×10^{12} potential HEAs with five elements from the set of fifty can be enumerated using a step of 1 at %. To date, only a few thousand different HCMAs have been produced worldwide, only a fraction of which are reported as HEAs in the scientific literature [11], and efforts have focused on an even more limited number of families [4]: in all likelihood, a huge number of HEAs remains to be discovered. One may further consider that the field is so recent that existing HEAs are most likely far from being optimised and that improvement margins have yet to be filled.

The compositional optimisation of HEAs is a task that should be possible to address using genetic algorithms, a class of evolutionary algorithms suited for maximising or minimising outputs over huge input spaces. Genetic algorithms have proved to be efficient in tackling complex optimisation problems in metallurgy, for instance for the design of steels [22,23] and superalloys [24,25]. Optimisation strategies usually consist in maximising or minimising a given calculated target — an "objective" — while obeying a number of constraints. The outcome is therefore a unique solution assumed to be the best in view of the sole objective. Besides, multi-objective genetic algorithms allow the concurrent optimisation of several target properties — while still possibly satisfying some constraints — so that several optimised solutions are provided, each being better than all others regarding at least one objective. For instance, a multi-objective optimisation scheme maximising strength and minimising density would produce several alloys, each of which is the lightest for its strength, and the strongest for its density. These alloys form the so-called Pareto set, which provides the decision-maker more freedom in the selection of an alloy in light of envisioned applications compared to mono-objective optimisation. Multi-objective genetic algorithms have also been applied to alloy design [26–29]. Our aim here is to perform a multi-objective compositional optimisation of HEAs using a genetic algorithm carrying out the simultaneous maximisation of strength and solid solution stability, as predicted using recent approaches [20,21], and minimisation of density. A preliminary experimental investigation is then performed in order to assess the proposed approach.

This exploration method thus builds on those of Miracle et al. [16] and Senkov et al. [17,18], who proposed tens of thousands of new potential equimolar HEAs by systematically estimating some of their properties using rules of mixture models and CALPHAD predictions. In contrast, the proposed method is designed for efficiently screening non-equimolar compositions. This means exploring a dauntingly larger space: for example, while there might be only 5005 six-element families (and thus equimolar alloys) given the number of elements chosen latter in this paper, every family "contains" 14 millions possible HEAs using an accuracy of 1 at %.

2. Alloy optimisation strategy

The alloy design strategy involves a multi-objective optimisation genetic algorithm which first needs to be specifically adapted to the design of HEAs, due to their particular combinatorial features. These adaptations are first presented in Section 2.1. The optimisation routine notably aims at maximising phase stability, which is evaluated by a novel statistical criterion defined in Section 2.2. Sections 2.3, 2.4 and 2.5 offer complementary insights into the design strategy regarding the estimation of density, SSH and crystal structure, as well as the role of computational thermodynamics.

2.1. Genetic algorithm and combinatorial issues

The design methodology makes use of a modified version of the popular multi-objective genetic algorithm NSGA-II [30]. The main principles of the algorithm have been exposed elsewhere [29] and will be briefly summarised here. Similarly to other genetic algorithms, the original NSGA-II works with a population of individuals — here symbolising alloys — over several generations. These individuals are defined as a combination of genes each corresponding to the content of an alloying element. At each generation, the genes may be crossed and mutated in order to modify the properties of the alloys in the population. At the end of a generation, the Darwinian "survival of the fittest" principle is applied, ensuring that the "best" alloys are kept in the population for the next generation, while the poorly performing alloys are rejected. A gradual improvement in performance is achieved through the iteration of this process, typically over hundreds or thousands of generations. This original genetic algorithm was modified [29], notably through the addition of a secondary population called "archive". Immutable and of unlimited size, the archive is updated at the end of every generation so that it stores only the Pareto set of HEAs found by the algorithm since the beginning of evolution. This allows the primary population to freely roam the search space, whose extent is such that even Pareto-optimal alloys might be dropped out of this population of limited size. For details on the inner working of the NSGA-II algorithm, and on the modifications brought to it, the reader is referred to the original papers [29,30].

Owing to their definition, high entropy alloys possess no proper base element: the preponderance of an element over the others cannot extend to that of a balance element in common alloy systems such as steels and nickel- or cobalt-based superalloys. When designing the latter, alloying elements added to the base element are the