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New strategies and tests to accelerate discovery and development of multi-principal element structural alloys

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ABSTRACT

High throughput, combinatorial approaches dramatically accelerate the discovery and development of functional materials, where properties depend primarily on composition. In structural materials, many properties depend sensitively on sample dimensions and microstructural length scales, complicating sample miniaturization that is critical to high throughput techniques. As a result, high throughput tests to rapidly evaluate structural materials are not currently available to meet the challenges offered by the extreme numbers of alloy compositions and microstructures. Here we develop a strategy to accelerate the exploration of conventional and multi-principle element structural alloys, and describe new tests for the rapid evaluation of structural alloys.

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1. Introduction

High entropy alloys (HEAs)[1] and the related concepts of multi-principle element alloys (MPEAs)[1] and complex, concentrated alloys (CCAs)[1] open alloy development to a vast richness of complexity. There are three dimensions to this complexity: a vast number of alloy bases (unique combinations of principal elements); a large number of alloys within each base (variations in principal element concentrations plus changes in minor alloying elements and their concentrations); and almost endless microstructures for each alloy. Combinatorial equations give hundreds of millions of alloy bases [1] and hundreds of billions of unique alloys. These numbers are beyond our ability to comprehend – we struggle to conceive of characterizing one hundred million alloy systems using current methods. To put this in perspective, the MPEA field has evaluated an average of 10 new alloy systems each year [1]. At this rate, it will take over 17 million years to study all alloy systems containing three to six elements. Clearly, radical new approaches are needed.

High throughput, combinatorial experiments can quickly evaluate many alloys [2–4]. This approach uses materials libraries with composition gradients and rapid experimental techniques that can be *automated*, *parallelized* and *miniaturized*. Such techniques are well established for functional materials. In fact, essentially all high throughput experiments are used for functional materials. The reason is simple – high throughput experiments rely on miniaturizing the sample volume, and functional properties can be measured reliably on very small volumes. This is because functional materials properties do not depend strongly on microstructure.

Structural materials are different. Structural properties depend on both composition and microstructure. A microstructural specification includes the types of phases present as well as their sizes, shapes, volume fractions, distributions and crystallographic orientations. Defects are also an essential part of microstructures. Almost endless microstructures can be produced, even at a fixed composition. Further, structural properties measured on microscopic volumes can be dramatically different than bulk properties [5]. Microstructure and length scale are major barriers to the use of high throughput experiments for structural materials.

The Materials Genome Initiative (MGI) integrates computational tools, digital data and experimental tools [6], but relatively little has been published on high throughput experiments to measure structural material properties. This gives a compelling challenge to the scientific community for the rapid exploration of structural alloys. High throughput experiments for structural materials require a different way of thinking. These experiments are designed for rapid screening, not for collecting accurate design data or for developing mechanistic models. Tests that are simple, repeatable and measure a material response that

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[1] Following an earlier discussion [1], the term HEA is used when configurational entropy or single-phase, solid solution microstructures are important. Otherwise, the terms MPEA or CCA are used to evoke the vastness of compositions and microstructures with no limitations on the magnitude of configurational entropy or the type of microstructures formed.

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correlates with the property of interest are needed. The hardness test is a well-known example. It is fast, simple and repeatable and although it doesn't measure tensile strength it correlates well enough with this property to guide exploration. High throughput evaluations need to be at least 100 to 1000 times faster than current tests. It generally takes weeks or months to evaluate tensile properties of a single alloy with conventional techniques. In rough numbers, compressing six weeks of work into 1 h gives an acceleration of 1000×. Some progress is being made. In a narrow window of alloy compositions and microstructures, rates approaching 1 alloy per h have already been demonstrated [7].

High throughput experiments to measure mechanical properties are not enough. New materials libraries are also needed [1,8], and the speed with which these materials libraries can be produced is important. New strategies to use these tools are also needed. This paper describes new ideas for high throughput experiments and new strategies to integrate them with other components of high throughput evaluation of structural materials.

2. A new strategy defined by two principles

Structural materials evaluation usually begins with 'first-tier' properties such as strength and ductility, then 'second-tier' properties such as fatigue, creep and fracture toughness, and finally by measuring properties such as environmental resistance. This approach is effective when candidates are limited to a small number of alloy families with known relationships between composition, microstructure and properties. This pre-established knowledge is captured in the materials selection process pioneered by Ashby [9]. The same approach is used to explore entirely new structural alloys, even though the relationships between composition, microstructure and properties are unknown. This dramatically slows the search for new structural alloys, since the knowledge needed to guide alloy modifications has to be established along the way. Current MPEA research uses this approach, and is establishing relationships between composition, microstructure and mechanical properties in an iterative feedback loop [1]. New relationships are produced, but at a significant risk, since the alloy family being studied may eventually be found to lack a useful balance of properties.

A new strategy is needed to improve the chances of discovering impactful, new alloys with a full balance of useful properties when exploring a vast number of unknown alloys. Here we develop a strategy that starts with the principle that *the time and resources spent evaluating each candidate alloy have an inverse relationship to the number of candidates* (Fig. 1). At the beginning of the discovery process, when the numbers are vast, the amount of time spent considering each candidate is very small. As evaluation proceeds, the amount of time afforded to remaining candidates can increase. This strategy is also based on the principle that *all candidates are systematically evaluated by the same evaluation process and selection criteria*. This is a dramatic change from the MPEA field, where different alloys are selected for study using different criteria and may be evaluated using different tests.

3. Many properties are required – a failure in one is a failure in all

A successful structural material must possess many properties [10]. A 'spider plot' (Fig. 2) illustrates typical properties for structural alloys as a percentage improvement over required values along 12 axes. A property that just meets the requirement is at the 100% mark, improved properties are shown outside the bold requirements profile (>100%), and inadequate properties fall inside this profile (<100%). For most properties, higher values are properties improvements. For density, lower values are better, so that a density that is 20% lower than the requirement is plotted at 120%, since it gives a 20% improvement. Not all of these properties are needed for every structural application, and some applications may require properties not shown in Fig. 2.

Two alloys are illustrated in Fig. 2. Alloy A has significant improvements in density and environmental resistance, and meets or exceeds

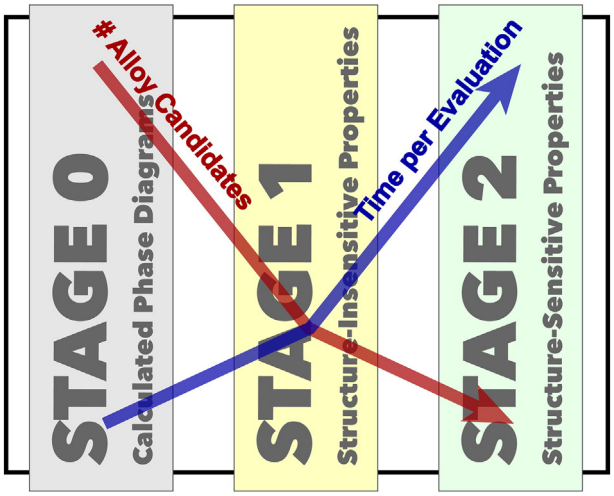

Fig. 1. Schematic illustration of the strategy proposed here. The first evaluation (Stage 0) performs high throughput calculations of phase diagrams that can give the desired microstructures. The first experimental step (Stage 1) evaluates properties that are insensitive to microstructure on materials libraries with controlled composition gradients. The final evaluation (Stage 2) measures high throughput properties that depend on both composition and microstructure using materials libraries of fixed composition and controlled microstructure gradients. The time spent evaluating each alloy is smallest when the number of candidates is largest and vice versa. Only alloys that pass a stage are considered in subsequent stages.

all other properties. Alloy A is a candidate for the target application. Alloy B has significantly higher yield and ultimate strength, as well as much improved creep and fatigue properties and environmental resistance. However, Alloy B does not meet the ductility requirement. Even though Alloy B gives significant improvements in several properties, it is not a candidate since it does not meet one of the required properties. This is a key point – a failure in one property signals a failure of the alloy as a whole.

In this paper, an alloy is defined by both composition and microstructure. This emphasizes that many properties aren't determined by composition alone and must include a description of the microstructure. Thus, while the ductility of Alloy B might be improved by changing the microstructure, this gives a new alloy with a new profile on the spider plot. The potential of new alloys is often evaluated by comparing a single

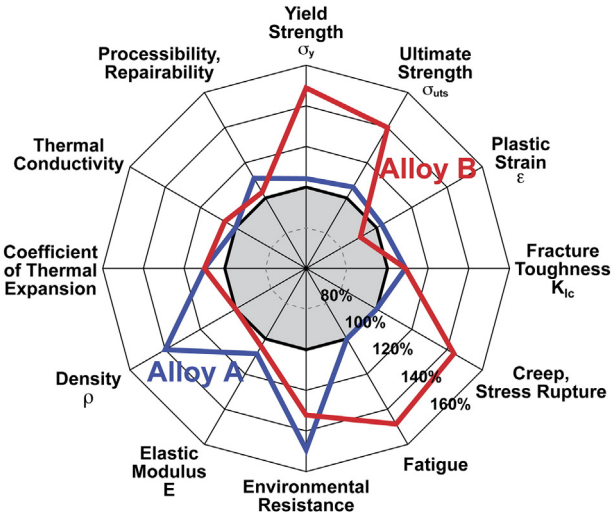

Fig. 2. A 'spider plot' illustrating properties that are typically required for structural materials. An alloy profile is constructed by plotting each alloy property relative to the required value on the 12 axes. A property that just meets the requirement falls on the bold profile at 100%, improved properties are shown outside of this bold profile (>100%), and inadequate properties fall inside the bold profile (<100%).

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