DEFINING PATHWAYS FOR REALIZING THE REVOLUTIONARY POTENTIAL OF HIGH ENTROPY ALLOYS

A TMS ACCELERATOR STUDY

TMS A Study Organized by The Minerals, Metals & Materials Society

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The Minerals, Metals & Materials Society (TMS)

Promoting the global science and engineering professions concerned with minerals, metals, and materials

The Minerals, Metals & Materials Society (TMS) is a member-driven, international organization dedicated to the science and engineering professions concerned with minerals, metals and materials. TMS includes approximately 13,000 professional and student members from more than 80 countries, representing industry, government and academia.

The society's technical focus spans a broad range—from minerals processing and primary metals production to basic research and the advanced applications of materials.

In recent years, TMS has established itself as a leader in advancing integrated computational materials engineering (ICME), computational materials science and engineering, multiscale materials modeling and simulation, materials data infrastructure issues, and advanced manufacturing methodologies.

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George Spanos, Project Leader

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Dan Miracle is a Senior Scientist in the Materials and Manufacturing Directorate of the Air Force Research Laboratory (AFRL). Miracle represents technologies of interest to the US Air Force and US Space Force and leads formation of technical partnerships within the government and with universities, industry, and the international scientific community. His research has covered nickel-based superalloys and intermetallic compounds; metal matrix composites; advanced aluminum alloys; and boron-modified titanium alloys. Miracle's current research explores metallic glasses and high entropy alloys.

Miracle received a B.S. degree in Materials Science and Engineering from Wright State University, M.S. and Ph.D. degrees in Metallurgical Engineering from The Ohio State University, and an Honorary Doctor of Science from the Institute of Metal Physics, Ukrainian Academy of Sciences. He is a Fellow of ASM, International; The Minerals, Metals & Materials Society (TMS); AFRL; and is an Honorary Member of the Indian Institute of Metals. Miracle received the AF Basic Research Award and the Presidential Rank Award. He is author or co-author of over 200 peer-reviewed scientific articles and seven book chapters, and he is co-editor of six books. Miracle has delivered more than 200 plenary, keynote, and invited talks.

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Donald Brenner received his B.S. from the State University of New York in 1982, and his Ph.D. from Penn State University in 1987, both in Chemistry. He then joined the research staff of the US Naval Research Laboratory as a member of the Theoretical Chemistry Section. In 1994, Brenner joined the faculty at NC State, where he is currently a Kobe Steel Distinguished Professor and Department Head in the Department of Materials Science and Engineering. His research focuses on the development and use of atomic, multi-scale, and statistical modeling methods for the virtual design, development, and characterization of advanced materials. His awards include the 2002 Feynman Prize for advances in nanotechnology, the 2013 Alcoa Foundation Distinguished Engineering Achievement Award, and the 2016 Alexander Quarles Holladay Medal for Excellence.

Andrew Detor
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Andrew Detor is a Principal Materials Scientist at GE Research with 12 years of experience developing and transitioning new technologies for GE businesses and government partners. Detor specializes in metallurgy, applying experimental and computational techniques to develop new alloys and processing routes aimed at improving system performance while satisfying customer requirements. His current work builds on a materials informatics framework, leveraging machine learning methods, integrated computational materials engineering (ICME), and multi-objective optimization to guide material discovery for ultrahigh-temperature use. Past projects have transitioned lab-scale development efforts to commercial application across a range of products, including large-scale gas turbine forgings, microelectromechanical systems, and corrosion resistant aircraft engine coatings. Current research interests lie at the intersection of high-throughput screening, knowledge representation, and 3rd wave artificial intelligence to advance materials science and bring new capabilities to market.

Prior to GE, Detor completed a postdoc at Lawrence Livermore National Laboratory, studying deposition methods to manufacture fuel capsules for inertial confinement fusion experiments. He earned his B.S. degree from Rensselaer Polytechnic Institute in 2003 and Ph.D. from Massachusetts Institute of Technology in 2007, both in Materials Science and Engineering.

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Katharine Flores received her Ph.D. in Materials Science and Engineering from Stanford University in 2000. After serving as a postdoc and the Director of the Sports Materials Laboratory at Stanford, she joined the Materials Science and Engineering faculty at the Ohio State University (OSU) in 2002. In 2008, Flores became the Director of Education and Outreach for Emergent Materials, the NSF Materials Research Science and Engineering Center (MRSEC) at OSU. In 2012, she moved to the Mechanical Engineering and Materials Science Department at Washington University in St. Louis, where she helped to establish the Institute of Materials Science and Engineering (IMSE) as its Associate Director. She became Director of the IMSE in 2016. In 2021, she served as the Interim Chair of the Energy, Environmental, and Chemical Engineering Department.

Flores' research focuses on structural materials, with particular emphasis on understanding structure-processing-property relationships in compositionally or structurally complex metallic alloys. Her current research projects include developing and applying high-throughput computational and experimental methods to alloy design and using micromechanical experimental methods to investigate the rheology of geological materials.

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Michael Gao is a physical scientist with Materials Engineering and Manufacturing Directorate at the Department of Energy National Energy Technology Laboratory (NETL). He received a Ph.D. degree in Materials Science from the University of Virginia (UVa) in 2002, followed by postdoctoral appointments at UVa and Carnegie Mellon University. Gao joined NETL as a contract researcher in 2008. His recent research focus is on accelerating the design and development of cost-effective, high-performance materials for extreme environments by integrating multi-scale computational modeling (including first-principles, molecular dynamics, Monte Carlo, finite element method, and CALPHAD) and machine learning with critical experiments. Gao has published more than 80 peer-reviewed journal papers and five book chapters; he holds one patent and has another one pending. Gao has co-organized many symposia and special issues on high entropy alloys at TMS and Materials Science & Technology (MS&T) conferences and in several journals. He coedited the book *High Entropy Alloys: Fundamentals and Applications* in 2016. He was the chair for the TMS Alloy Phases Committee (2017–2019). Currently, Gao serves as the vice chair of the Alloy Phase Diagram Committee of ASM International and a Principal Editor for the *Journal of Materials Research.* He received the best paper award in 2005 by the Alloy Phase Diagram International Commission.

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Easo George earned his B.Tech. in Metallurgical Engineering from the Indian Institute of Technology, Kanpur, in 1981, and Ph.D. in Materials Science and Engineering from the University of Pennsylvania in 1985. After brief stints as a Post-Doctoral Fellow at Penn and a Research Engineer at Southwest Research Institute in San Antonio, he joined the Oak Ridge National Laboratory (ORNL) in 1987 and rose to become Distinguished Research Staff Member and Head of the Alloy Behavior and Design Group. From 2002 onward, George was also a Joint Faculty Professor of Materials Science and Engineering at the University of Tennessee (UT). In 2014, he moved to Germany to become Professor of Materials Design and Director of the Center for Interface-Dominated High-Performance Materials at the Ruhr University in Bochum. In 2017, George returned to Tennessee, where he is currently the UT-ORNL Governor's Chair for Advanced Alloy Theory and Development.

George's research has revolved around the physical metallurgy and mechanical behavior of metallic materials. Most recently, it has focused on understanding phase stability and deformation mechanisms of high entropy alloys, areas in which George has published several highly cited papers. Prior to that, he made seminal contributions to small-scale mechanical behavior, alloying effects on deformation and fracture of iridium alloys, environmental embrittlement and alloying effects in nickel and iron aluminides, vacancy induced anomalous strengthening of iron aluminides, and the hierarchy of creep cavity nucleation sites in ferrous alloys.

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Chris Haines is currently a Senior Metallurgist with US Army DEVCOM – Army Research Laboratory at Aberdeen Proving Ground, MD. Haines works in the area of nanocrystalline materials and is focused on exploiting the unique properties of these materials for Army applications. His research has delved into nanoscale metallic particles for energetics, nanostructured metal alloys for structural applications, and nanostructured ceramics for armor. Haines has focused on both synthesis and processing of nanomaterials, including investigating field-assisted sintering as a technology for obtaining bulk, nanostructured materials. More recently, his focus is on demonstrating high-throughput materials science, including the integration of machine learning and artificial intelligence into the materials development cycle. To this end, Haines' team has begun employing high-throughput processes for developing refractory high entropy alloys (RHEA) for applications in hypersonics.

Haines has a B.S., M.S., and Ph.D. in Ceramic & Materials Science Engineering, all from Rutgers University. He previously worked at US Army Armaments Research & Development Engineering Center (ARDEC) at Picatinny Arsenal, NJ. Haines holds nine US Patents and is very active in STEM activities (local outreach, eCybermission, SMART scholarship panels, and the National Junior Science & Humanities Symposium).

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Keith Knipling received his Ph.D. in Materials Science and Engineering from Northwestern University in 2006, where his research was toward developing new precipitation-strengthened aluminum alloys for high-temperature applications. Knipling came to the US Naval Research Laboratory (NRL) in 2006 as a National Research Council Associate, where his research sought to understand the microstructural evolution and deformation mechanisms that occur during friction stir welding of a variety of structural alloys. Currently a staff scientist at NRL, Knipling's research centers on studying a variety of structural and functional materials on the nanoscale by atom probe tomography, including the design of new high-temperature aluminum alloys, developing more energy efficient soft magnetic alloys, and the discovery of new high entropy alloys for extreme environments. He is a 2008 and 2009 recipient of the NRL Alan Berman Research Publication Award and a 2012 recipient of the Presidential Early Career Award for Scientists and Engineers (PECASE), the highest honor bestowed by the United States government on outstanding scientists and engineers in the early stages of their independent research careers. He is a lifetime member of TMS and serves on the TMS Phase Transformations Committee.

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John Lewandowski is a Distinguished University Professor and the Arthur P. Armington Professor of Engineering at Case Western Reserve University. He is Director of the Advanced Manufacturing and Mechanical Reliability Center (AMMRC) http://ammrc.case.edu. His primary appointment is in Materials Science and Engineering with a secondary appointment in Mechanical and Aerospace Engineering. Lewandowski's BS/ME/PhD are in Metallurgical Engineering and Materials Science, earned at Carnegie Mellon University where he was a Hertz Foundation Fellow, followed by a NATO/NSF Postdoctoral Fellowship at Cambridge University. Lewandowski is also Overseas Fellow at Churchill College, Cambridge University and Visiting Professor at Nanyang Technological University. His publications/presentations exceed 345 and 1050, respectively, in areas of processing/structure/property relationships in crystalline and amorphous advanced materials systems for aerospace, automotive, biomedical, and defense applications. Lewandowski's recent work focuses on HEAs, as well as additive and other advanced manufacturing techniques. His collaborative work continues using synchrotron experiments to image damage evolution and environmental fracture in various structural aluminum alloys. Lewandowski's various national/international awards for research and teaching/mentoring activities include the TMS Leadership Award, 2022 ASM Albert Easton White Distinguished Teacher Award, ASMI Fellow, and the Inst. of Metals Charles Hatchett Award for work on Nb. He has served on numerous NSF and NAS/NRC panels while also serving on the editorial board of a number of journals.

Tresa Pollock
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Tresa Pollock is the Alcoa Distinguished Professor of Materials and Associate Dean of Engineering at the University of California, Santa Barbara. Her research focuses on the mechanical and environmental performance of materials in extreme environments, unique high temperature materials processing paths, ultrafast laser-material interactions, alloy design, and 3-D materials characterization. Pollock graduated with a B.S. from Purdue University in 1984 and a Ph.D. from MIT in 1989. She was employed at General Electric Aircraft Engines from 1989 to 1991, where she conducted research and development on high-temperature alloys for aircraft turbine engines and co-developed the single crystal alloy René N6 (now in service). Pollock was a professor in the Department of Materials Science and Engineering at Carnegie Mellon University from 1991 to 1999 and the University of Michigan from 2000–2010. Her recent research has focused on development of new femtosecond laser-aided 3-D tomography techniques, damage detection and modeling by resonant ultrasound spectroscopy, thermal barrier coatings systems, new intermetallic-containing cobalt-base materials, nickel base alloys for turbine engines, multi-principal element alloys, and bulk nanolaminates. Pollock was elected to the US National Academy of Engineering in 2005, the German Academy of Sciences Leopoldina in 2015, and is a DoD Vannevar Bush Fellow and Fellow of TMS and ASM International. She serves as editor-in-chief of the Metallurgical and Materials Transactions family of journals and was the 2005–2006 president of The Minerals, Metals & Materials Society.

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John R. Scully is the Charles Henderson Chaired Professor of Materials Science and Engineering (MSE), the Co-director of the Center for Electrochemical Science and Engineering, and the MSE Department Head at the University of Virginia. Prior appointments were at Sandia National Laboratories and the Naval Ship Research and Development Center. Scully received a Ph.D. in MSE from the Johns Hopkins University. His field of research is corrosion, focusing on circumstances where metallurgical factors such as structure, composition, and processing are important governing factors. His research and education focus on structural and functional materials as well as coatings, often combining in situ and ex situ test methods. Electrochemistry, surface science, and materials science are frequently integrated. He has co-authored numerous scholarly publications in these areas. Scully is a Fellow of the Electrochemical Society, the National Association of Corrosion Engineers, The British Corrosion Institute, and ASM International. He served on the editorial boards of *Materials and Corrosion,* and *Metallurgical Transactions*. He is the Chief Technical Editor of *CORROSION*, *The Journal of Science and Engineering*. He has contributed to several National Academy, Defense Science Board Task Force, and international corrosion reviews, including the review of the corrosion of spent nuclear fuel containers in five countries. He has received numerous awards such as the British Corrosion Institute U.R. Evans Award, the Whitney Award from NACE, and the H. Uhlig Award from ECS and NACE.

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Alejandro Strachan is a Professor of Materials Engineering at Purdue University, Director, DoD ONR MURI "Predictive Chemistry and Physics at Extreme Conditions" (PCP@Xtreme), and the Deputy Director of NSF's nanoHUB. Before joining Purdue, Strachan was a Staff Member in the Theoretical Division of Los Alamos National Laboratory and worked as a Postdoctoral Scholar and Scientist at Caltech. He received a Ph.D. in Physics from the University of Buenos Aires, Argentina. Strachan's research focuses on the development of predictive atomistic and multiscale models to describe materials from first principles and their combination with data science to address problems of technological or scientific importance. Areas of interest include high-energy density and active materials, metallic alloys for high-temperature applications, materials and devices for nanoelectronics and energy, as well as polymers and their composites. Strachan has published over 180 peer-reviewed scientific papers and his contributions to research and education have been recognized by several awards, including the Early Career Faculty Fellow Award from TMS in 2009, his induction as a Purdue University Faculty Scholar (2012–2017), and the R&D 100 in the area of software and services for nanoHUB.

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Mitra Taheri is a Professor of Materials Science and Engineering and the Director of the Materials Characterization and Processing (MCP) Center at Johns Hopkins University (JHU). Prior to her position at JHU, Taheri held the Hoeganaes Endowed Chair Professorship in the Department of Materials Science & Engineering at Drexel University. Taheri leads the Dynamic Characterization Group, focusing on the development and use of innovative in situ microscopy and spectroscopy to characterize microstructural evolution and properties of materials in a wide variety of environments and external stimuli. Her current research focuses on the following topics: instrumentation development for electron microscopy and spectroscopy; machine learning; high-throughput methodologies; physical metallurgy and in particular, understanding microstructural evolution; materials processing; metal additive manufacturing (3D printing); magnetic composite development; optimization and in situ processing of quantum materials, 2D materials, and surfaces; and biomaterials, with a particular focus on maternal fetal medicine.

Taheri obtained her Ph.D. from Carnegie Mellon University, followed by an NRC Postdoctoral Fellowship at the Naval Research Laboratory (NRL) and a Director's Postdoctoral Fellowship at Lawrence Livermore National Laboratory (LLNL). During her doctoral studies, Taheri was a visiting scholar at RWTH Aachen University, the National Center for Electron Microscopy (LBL), and Northwestern University's Center for Atom Probe Tomography. She has received numerous awards, including but not limited to the NSF Career Award, DOE Early Career Award, ONR Summer Faculty Fellowship, Engineers As Global Leaders for Energy Sustainability ("EAGLES") Faculty Mobility Fellowship (at Politecnico di Milano), Microscopy Innovation Award, R&D 100 Award, US Steel Graduate Fellowship, MRS Graduate Student Award, and induction to Sigma Xi. Taheri has published ~150 peer-reviewed articles in journals such as *Science*, *Nature Communications*, *ACS Nano*, *Nanoletters*, and *Acta Materialia*, and has garnered ~175 invited and keynote presentations and seminars across the world.

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Michael Titus is an Assistant Professor of Materials Engineering at Purdue University. Prior to joining Purdue, Titus earned his Ph.D. in Materials from the University of California, Santa Barbara in 2015. His research focuses on accelerating design of structural alloys for high-temperature applications by integrating high-throughput experiments with thermodynamic and atomistic simulations via data science. His other research interests include understanding solute segregation to nanocrystalline defects in alloys by a combination of first-principles calculations and high-resolution characterization methods and exploiting these atomic-scale processes to improve the high-temperature mechanical properties of structural alloys. Titus has co-authored more than 25 peer-reviewed papers. In 2019, he was awarded the National Science Foundation's CAREER award, and recently in 2020, he was recognized as a TMS Young Leaders International Scholar with the Japanese Institute of Metals and Materials.

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The Boeing Company

Ali Yousefiani is a Boeing Technical Fellow and the Chief Scientist for Metallic Materials & Manufacturing Technology within Boeing Research & Technology. In this role, he helps lead the team's strategic investment decisions and enterprise level R&D initiatives. Yousefiani is also responsible for program execution and insertion of advanced/disruptive metallic-based structural materials technology solutions into current and future Boeing product platforms. His key areas of emphasis include predicting behavior of materials and structures in extreme environments associated with hypersonic flight, atmospheric reentry, rocket/aircraft propulsion, and integrated power/thermal management; and research and development of novel performance enhanced materials and their powder-based methods of processing and production.

Yousefiani currently leads programs aimed at development and maturation of manufacturable, durable, and rapidly deployable multifunctional metallic airframe structures for Boeing's extreme environment applications, and development of extreme environment heat exchangers and ultra-high performance turbine blades made from novel complex concentrated / high entropy alloys produced using traditional and additive manufacturing methods. Yousefiani received his Ph.D. in Materials Science and Engineering from the University of California, Irvine in 1999 and continues to lecture in the School of Engineering. He has 30 years of experience in the field.

Final Report Review Team

Following the writing and editing of multiple drafts of this report by the TMS science and engineering staff and the lead expert study team, an independent review team provided valuable detailed comments and suggestions for implementation into the final report. Their efforts are gratefully acknowledged, and this team included:

- **Irene Beyerlein**, University of California, Santa Barbara
- **Nick Birbilis**, The Australian National University
- **Krishanu Biswas**, Indian Institute of Technology (IIT) Kanpur
- **Todd Butler**, Air Force Research Laboratory
- **Amy Clarke**, Colorado School of Mines
- **Amy Gandy**, University of Sheffield
- **Nicholas Jones**, University of Cambridge
- **Eun Soo Park**, Seoul National University
- **John Sharon**, Raytheon Technologies

Other Key Contributors

This study and report were undertaken on behalf of the Defense Advanced Research Projects Agency (DARPA), from whom funding support is gratefully acknowledged, and the Air Force Research Laboratory (AFRL), and executed by The Minerals, Metals & Materials Society (TMS). The principal investigator of this activity was George Spanos, Director of New Initiatives, Science, and Engineering at TMS. The technical experts (see above) were convened, and their input was compiled into the final report through the efforts of TMS and Nexight Group, LLC. Other important TMS staff contributions to this report include those of Michael Rawlings (Science and Engineering Lead), Amy DeFilippo (Technical Project Administrator), Kelly Markel (Publications Coordinator), Bob Demmler (Graphic Designer), Marleen Schrader (Accounting and Human Resources Specialist), Ann Ritchie (Technical Communications Specialist), David Rasel (Media Manager), Lynne Robinson (Department Head, Strategic Communications and Outreach), Matt Baker (Department Head, Content), Annie Ooi (Technical Intern), and Zachary Trdinich (Science and Technology Intern). Nexight Group, LLC staff members who were heavily involved in this effort include Ross Brindle (Chief Executive Officer) and Jared Kosters (Senior Technical Consultant).

Preface

Why study high entropy alloys?

High Entropy Alloys (HEAs) represent a fundamental divergence from the way humankind has approached alloy development for the last 5,000 years. Instead of starting with one base element which accounts for most of the alloy composition and adding dilute amounts of other elements, HEAs focus on the unexplored central regions of multi-element phase diagrams, where three or more alloying elements occur in concentrated amounts, and there is no obvious single base element. This novel approach opens the door to millions of new alloy systems, including materials with unique, never-before-seen combinations of structural and/or functional properties. Recently developed HEAs have demonstrated strong potential to impact many applications of interests, such as those in defense-related domains, through their exceptional high-temperature, light-weighting, corrosion resistance, and radiation resistance properties, amongst others. If properly developed, HEAs have the potential to provide revolutionary new capabilities. This report delves deeply into the background, value, underlying technologies, challenges, and opportunities associated with HEAs. It then provides detailed recommendations and actionable tactics to help accelerate the discovery, development, and implementation of these promising materials. As this study has been supported by the US Defense Advanced Research Project Agency (DARPA), special focus is given to defense applications.

Who should read this report and why?

This report should be of interest to researchers, engineers, technical policy leaders, and those who are involved in technology development for national security, economic, and/or societal interests. More specifically, this report contains detailed information, analysis, and recommendations regarding this potentially revolutionary materials development approach which should be of high value to scientists, engineers, and designers within materials and manufacturing communities, as well as those within several related disciplines, such as computer and data science, physical chemistry, and/or mechanical engineering. These technical experts are expected to span academic, industrial, and governmental sectors.

This report should also be of use to leaders and decision makers in the US Department of Defense (DoD) and other branches of the government and industry, who will especially benefit from the assessment of current technical challenges and the recommended action plans provided to accelerate the development of HEAs. More specifically, the DoD, other federal agencies, private entities, national initiatives, or other institutions that support or fund the development or production of manufactured functional or structural parts should find this report to be useful. Other groups that may also be interested in the contents of this report include (1) policymakers at the local, state, and federal levels, (2) educators teaching undergraduate and graduate courses on materials engineering, manufacturing, and/or advanced computation, (3) industry lead technologists in charge of road mapping novel materials and manufacturing technologies for critical applications, and (4) department heads and/or deans looking to advance the curriculum around these topical areas. Since much of this report relates to effectively and efficiently discovering, developing, and deploying the next-generation of high-performance materials— particularly within the materials science and engineering (MSE) and manufacturing communities —individuals and organizations who influence the futures of these communities may benefit by taking advantage of the insights from this study.

How to navigate this report

Readers are encouraged to navigate this report by first examining the Executive Summary for an overview of the structure and highlights of this document and to determine which parts might be of most relevance to your expertise, interests, and/or organization. It is our hope that this report will inspire you to take specific actions consistent with your skills and interests to support the discovery, development, and eventual widespread deployment of novel HEAs in numerous critical applications. The Introduction and Further Background and Study Motivation sections provide insight into the current landscape and history of this emerging field, while the Value Proposition and Target Application Areas sections articulate some of the potential advantages and applications of HEAs. The Challenges, Needs, and Limitations section is meant to prompt you and your colleagues to think critically about the challenges that, if overcome, will most affect rapid development of HEAs, and to identify the challenges and opportunities to which you may be able to contribute solutions and progress. In both the Preliminary Recommendations and Action Plans sections, you will find suggested actionable next steps and detailed tactics to overcome barriers and accelerate development and implementation of HEAs.

Hopefully, as you explore these sections, you will begin to focus on the tactical details that resonate most with your interests and expertise and will prioritize some specific actions that you and your colleagues might undertake.

Call to Action: What action should be taken after reading this report?

A major goal of this study is to stimulate direct action by a wide variety of stakeholders. Such actions should be centered on supporting acceleration of the discovery, development, and/or implementation of these potentially revolutionary new materials. After reading this report, some general next steps could include: (1) identifying specific challenges or recommendations that you and your colleagues could address, and from which the most benefit would be gained, (2) sketching out a detailed personal or organizational action plan, and (3) taking concrete steps to initiate this activity. These steps would be different depending on your role and area(s) of interest.

The HEA field is highly interdisciplinary, and many experts from the MSE and manufacturing communities will be needed to address the numerous challenges, opportunities, and recommendations presented in this report. It will also be critical to engage others beyond MSE and manufacturing in this discussion, including experts in the fields of computer science, data sciences and informatics, physics, chemistry, mechanical engineering, and multidisciplinary design optimization, to name a few. Identifying and establishing effective interdisciplinary collaborations will be a vital part of realizing the full potential of HEAs. The specific recommendations and action plans identified within this report should in no way be viewed as all-inclusive, and the leaders, researchers, engineers, and policymakers who read it are encouraged to develop and execute other activities and tactics as well. Our desire is that the readers will act promptly on the recommendations of this report. Although much work remains to be done, the potential is great for making impactful, short-to-medium-term progress on the discovery, development, and implementation of HEAs, as well as foundational, longer-term contributions to this potentially game-changing materials design approach.

Executive Summary

Background, Motivation, and Study Process

High Entropy Alloys (HEAs) represent a departure from the classic alloy design approach in which one base element accounts for most of the composition of the material. Instead, HEAs focus on the mostly unexplored central regions of multi-element phase diagrams, where there is no obvious base element. This results in hundreds of billions of possible HEA compositions. Moreover, HEAs have demonstrated unique and promising properties, providing potential for greatly enhanced performance in a variety of applications.

In order to help equip the materials science and engineering (MSE) and related communities to take advantage of the revolutionary potential of HEAs, this science and technology accelerator study aims to: (1) highlight the current state of HEA-related research; (2) scope and prioritize application and alloy domains of most promise (particularly for defense-related applications); (3) identify the unique needs and key enablers of next-generation HEAs; and (4) provide recommendations and action plans that will result in significant progress within the next 3 to 5 years toward breakthroughs in the scientific exploration, engineering development, and industrial implementation of HEAs. A team of internationally renowned experts was assembled for this study to achieve these objectives. Their inputs were collected at several live, interactive, virtual workshop sessions, as well as a series of online meetings and homework assignments, and synthesized into this report.

HEA Value Proposition

As with many new and possibly transformative materials and/or technologies, a strong value proposition must be clearly articulated, since early-stage innovations require significant resources and cultural "buy-in" to achieve their highest potential. This is particularly true of HEAs as they represent a departure from the millennia-old traditional methods of materials/alloy development. Section III considers the value proposition for HEAs in depth, from which the key benefits are summarized below.

Table 1. Value Proposition: Thematic Areas and Key Benefits

Thematic Area	Key Benefits
Expansive array of potential new high-performance HEA materials	New and unconventional property combinations
	Candidates for high-temperature applications
	Potential for key energy-related applications
	Potential for functionally graded properties
	High return on investment for materials discovery and development infrastructure
New cost-effective HEA systems and processing approaches	Lower-cost material substitutes
	Decreased reliance on critical materials
	New processing means and/or recycling methods
	Leverage ongoing DoD interest and support in additive manufacturing technologies
	Existing industrial infrastructure advantages
Advantages associated with leveraging data-driven materials discovery and design	Vast compositional design space and associated data
	ICME-derived HEA designs
	Machine learning (ML) -accelerated discovery of HEAs
	Contributions toward developing a new era of characterization tools
Vehicle to develop a highly competitive materials science and engineering workforce	Next-generation Materials Genome Initiative (MGI) workforce
	Improved readiness of existing workforce

Target Application Areas

Building upon the value proposition, various applications for which newly developed HEA materials might make a substantial impact are determined and discussed in some depth in Section IV, with a particular focus on potential DoD applications. The high-level synopsized list of target application areas is reproduced below:

1. Refractory HEAs (RHEAs)
2. Ultrahigh-temperature ceramics (e.g., for turbines)
3. Catalysts
4. Corrosion-resistant coatings
5. Light-weight materials
6. Novelty HEA systems
7. Thermal barrier coatings (TBCs)
8. Cryogenic systems
9. Batteries/superconductors
10. Thermoelectric materials
11. Electromagnetic applications
12. Anti-bacterial properties/applications
13. HEAs to enable renewable/alternative fuels
14. Quantum computing materials
15. Hydrogen-compatible/storage materials
16. High-entropy brasses and bronzes

Challenges, Needs, and Limitations

Table 2 summarizes the most significant challenge areas, needs, and limitations currently preventing more rapid development, emergence, and implementation of HEAs, as discussed in depth in Section V.

Table 2: Key challenge areas and supporting needs/limitations (reproduced from Section V).

Key Challenge Area	Needs/Current Limitations
A. High-Throughput Screening Methods and Experimental Tools	• High-throughput, automated, and/or autonomous tools and processes for integrated synthesis, characterization, and evaluation of HEAs • High-throughput experimental approaches for melting temperature; tensile strength and ductility; and toughness • High-throughput surrogate experiments for expensive and/or slow tests
B. Predictive Models and Computational Tools	• Fundamental theory for complex compositional space • Uncertainty-based predictive computational models for HEA development • Accurate cross-potentials for computational models • Computational tools for predicting structural and functional properties • ML approaches to help guide alloy selection • Visualization tools for interpreting complex phase spaces
C. High-Temperature Equipment and Testing	• High-temperature processing, testing, and property measurements • Addressing simultaneously the constraints of processing conditions (heating and oxidation) and sample size for high-temperature testing • Methods to process high melting point (e.g., >2000°C) RHEAs • Robust high-temperature die materials
D. Scattered Data with Uncertain Materials Pedigree	• Robust, coordinated, pedigreed datasets to supplant the disparate current data across the wide spectrum of HEA compositions • Widely adopted schema to establish provenance for HEA metadata
E. Fundamental Composition-Processing-Microstructure-Properties Knowledge	• Enhanced composition-processing-microstructure-properties correlations with as broad an applicability range as possible
F. In Situ Characterization Methods	• In situ monitoring and characterization tools to track all test parameters • Ability to monitor microstructural evolution in situ
G. Thermodynamic Databases	• Publicly available thermodynamic databases for HEAs • Consistency across methods used to gather HEA data • Multicomponent data to extrapolate into un-explored space • Entropy properties (in thermodynamic databases) that are efficient and flexible
H. Availability of Affordable Powder	• Solutions to overcome prohibitively expensive HEA raw material costs
I. Workforce Trained in HEA Exploration and/or Development	• A workforce skilled in using experimental and/or computational approaches and tools geared toward HEA exploration and development

Preliminary Recommendations

Section VI presents and discusses in some depth the preliminary recommendations to address the challenges and needs discussed above. These preliminary recommendations are grouped within nine high-level challenge areas and are summarized below.

A. High-Throughput Screening Methods and Experimental Tools

1. Develop a set of high-throughput HEA test methods for measuring properties of refractory HEAs (RHEAs) and ultrahigh-temperature ceramic HEAs
2. Develop a closed-loop autonomous screening capability that integrates materials synthesis, characterization, and machine learning (ML) computational tools
3. Devise new strategies for high-throughput experiments and computations
4. Increase coordination among government agencies
5. Establish a DoD-led consortium group to prioritize, generate, and share pre-competitive HEA data
6. Develop experimental tools for time-dependent material properties
7. Develop a high-throughput processing approach for bulk HEA samples
8. Create a mechanism for using shared high-throughput testing resources
9. Explore novel processing approaches for RHEAs
10. Develop a facility to conduct parallel high-throughput tests for strength, toughness, and creep
11. Establish data management and mining approaches

B. Predictive Structure-Property Models and Computational Tools

12. Generate an HEA structure-property knowledge base
13. Construct a set of reduced order models to help rapidly down-select HEA compositions
14. Develop reduced order models to study creep behavior
15. Develop and/or enhance oxidation models
16. Develop strengthening models
17. Develop thermodynamic models for the accurate prediction of phase stability and equilibrium
18. Develop tools for calculating thermal, phonon, and electron conductivities
19. Code a new suite of ML algorithms designed for HEA discovery and design
20. Develop predictive tensile and high-temperature property computational models designed specifically for corresponding high-throughput HEA test methods
21. Develop and validate, with uncertainty quantification (UQ), first-principles predictive models for HEA behavior in extreme conditions

C. High-Temperature Testing and Processing Equipment

22. Establish rapid screening methods for HEAs in high-temperature environments (≥1300°C)
23. Develop standards for inductive and resistive ultrahigh-temperature (UHT) testing

D. Scattered Data with Uncertain Materials Pedigree

24. Convene a broad HEA data consortium or working group to coordinate approaches to key data issues
25. Identify high-priority HEA systems to generate substantive pedigreed materials information

E. Fundamental Composition-Processing-Microstructure-Property Knowledge

26. Use artificial intelligence (AI) algorithms and other data informatics approaches to support composition-processing-microstructure-property relationships
27. Expand knowledge of new HEA systems and processing conditions

F. In Situ Characterization Methods

28. Designate national user facilities

G. Thermodynamic Databases for Complex Concentrated Alloy Combinations

29. Develop an open-source CALPHAD database for new HEA systems
30. Develop a data schema framework around thermodynamic databases
31. Populate new HEA thermodynamic databases using experimentally established phase equilibria data for concentrated multi-component alloys

H. Availability of Affordable Powder

32. Establish a research-scale powder manufacturing facility for HEA development
33. Form a centralized feedstock facility or consortium group

I. Workforce Trained in HEA Exploration and/or Development

34. Establish graduate-level internships and cooperative educational programs
35. Create HEA-related tutorials
36. Produce a multi-institutional workshop series

Action Plans

Using the challenges and needs (Section V) and preliminary recommendations (Section VI) as a foundation, a set of five high-priority action plans were developed, along with detailed tasks/activities for each action plan. These action plans and tasks (from Section VII) are synopsized below. Issues discussed in depth in Section VII for each of the 38 recommended tasks below include: time frame for completion, milestones and/or progress metrics, estimated costs, and the types of key players and roles required. The intent of these recommended action plans and tasks is to help scientists, engineers, and other stakeholders interested in HEAs accelerate the discovery, development, and implementation of these potentially game-changing materials.

Action Plan 1:
Develop High-Throughput Evaluation and Testing Methods

1.1 Develop a search strategy that maximizes information from a minimal set of evaluations
1.2 Develop new synthesis methodologies for establishing HEA materials libraries
1.3 Develop post-synthesis thermomechanical processing techniques
1.4 Design an autonomous materials research (AMR) Platform for HEAs
1.5 Establish high-temperature, high-throughput tests

1.6 Establish a database and common data schema and infrastructure for high-throughput HEA data
1.7 Establish active (extrapolative) machine learning methods
1.8 Integration and demonstration

Action Plan 2:

Develop or Extend Foundational Theory and Computational Models for HEAs

2.1. Develop or extend theory to accommodate multiple major elements
2.2. Extend theory and modeling of surface/subsurface thermodynamics and kinetics
2.3. Develop interatomic potentials for large-scale molecular dynamics (MD) simulations
2.4. Develop ability to predict electron-phonon scattering lengths
2.5. Develop computational models for predicting behavior under extreme conditions
2.6. Generate input data for ML models
2.7. Develop validated ML models for phase and properties predictions
2.8. Develop phase-field modeling to simulate corrosion and oxidation

Action Plan 3:

Develop a Nationwide Network of Interconnected HEA Partnerships

3.1. Form a government agency working group, team, or oversight committee
3.2. Establish the incipient national network of HEA Centers of Excellence (CoEs)
3.3. Develop strong academic–industry–Government partnerships for HEAs
3.4. Launch a DoD-led consortium on high-throughput/autonomous research
3.5. Assign specific HEA topics to future CoEs
3.6. Launch an HEA CoE for theory, modeling, and data acquisition/management strategies
3.7. Launch an HEA CoE for technology transfer and scale-up
3.8. Launch an HEA CoE for design-quality test data collection, storage, analysis, and sharing

Action Plan 4:

Engage in Feedstock Production and Scalability Research

4.1. Establish a small-batch production facility
4.2. Identify existing powder production facility partners to aggregate feedstock demand
4.3. Introduce material provenance requirements for property databases
4.4. Provide researchers access to thermomechanical processing facilities
4.5. Research new powder production methods
4.6. Establish high-throughput screening protocols to access HEA processibility
4.7. Generate processing and property data for legacy and emerging HEA materials
4.8. Develop melt-less processes for ultrahigh-temperature materials
4.9. Conduct research on powder recycling issues

Action Plan 5:

Develop Training and Resources for Workforce Readiness

5.1. Organize courses and/or workshops on applicable computational techniques
5.2. Identify internship opportunities
5.3. Incorporate HEA topical foci into Materials Science & Engineering (MSE) curricula
5.4. Develop HEA textbooks and/or chapters
5.5. Create certificates and/or credentials associated with short courses

I.
Introduction

Roughly 5,000 years ago, the first alloy developers observed that diluting the base metal copper with small additions of tin created a material with significantly improved strength, bronze. This simple discovery changed the course of human civilization and launched the Bronze Age. Subsequently, alloying iron with controlled, small carbon additions created steel, sparking the Iron Age and setting the groundwork for the Industrial Revolution. In the 20th century, as more metallic base elements were discovered, alloying led to the development of lightweight and high-temperature materials essential for "new aerospace industries, fueling additional societal transformations through air transportation, communication and navigation via satellites and GPS."[1] Although alloying techniques have become increasingly complex and sophisticated,[2] the basic concept of lightly modifying a base element with minor amounts of other elements to improve the properties currently remains as the dominant approach to alloy development. However, there are signs that this millennia-old approach may be reaching its limits. Researchers have thoroughly studied and exploited all the best metallic elements for engineering applications and have not discovered new stable metallic elements within the past century. The lack of novel metallic base elements makes it both challenging and expensive to develop new materials to meet the new generation of societal challenges using the lightly-alloyed-base-element method.

High Entropy Alloys (HEAs) represent a fundamentally different approach to alloy development. Instead of relying on a single base element that accounts for most of the final alloy composition, HEAs focus on the unexplored central regions of multi-element phase diagrams, where many alloy elements are concentrated and there is no obvious base element. Upon their discovery in 2004,[3–5] HEAs were defined as composed of five or more principal elements in (near) equimolar ratios. Due to their high configurational entropy, researchers focused on single-phase, disordered solid solution metallic alloys. Additionally, HEAs are considered a sub-component of multi-principal element (MPE) alloys, which were also discovered in 2004.[6] Since then, significant scientific exploration and tremendous growth expanded the definition and scope of HEA research to now include materials with as few as 3 principal elements where the maximum element concentration may be higher than 35 atom percent, intermetallic and ceramic compounds, as well as microstructures with any number and any type of phases.[7] New terminology such as complex concentrated alloys (CCAs) and high entropy materials (HEMs) is starting to emerge to embrace this broader research scope. Complex concentrated alloys (CCAs) include all alloys in the HEA field as well as other alloys over the vast number of compositions and microstructures in the central regions of multi-component phase diagrams.[2]

The HEA field has created, on average, nearly one new combination of elements and 3 new alloys (different elemental concentrations within each combination of elements) each month over the past 15 years.[7] With the rate of new systems of interest being discovered, it is challenging to fully explore the enormous number of HEA compositions and their microstructures, not to mention the extent of their potentially advantageous properties and applications. Moreover, every HEA represents a new, potential "base" alloy, since each HEA can be modified by minor elemental additions in a similar manner to traditional base alloys.[1] Varying the specific principal elements and compositions could unlock hundreds of billions of different HEA "base" alloys,[1] providing an opportunity to design new materials with properties tailored to the distinct needs of given applications. Thus, HEAs have great potential in a variety of domains, especially within defense-related applications. For example, HEAs could be designed to withstand higher operating temperatures in order to significantly improve operational windows in aerospace and various turbine applications, and increase thermodynamic efficiency.[8,9] HEAs also hold great promise for light-weighting in aerospace, ground vehicle, and/or maritime applications, resulting in improved performance and cost savings (e.g., vehicle speed and/or fuel savings). As one specific example, researchers determined that HEA alloy Al20Be20Fe10Si15Ti35 offers more than two times the specific strength of most commonly used titanium alloys in aerospace and maritime applications (e.g., Ti-6Al-4V).[8,9] HEAs offer potential breakthrough applications in hypersonics, corrosion-resistant alloys[10] (e.g., for Naval platforms and other harsh environments), and radiation resistance.[8,11] These are just some examples of the potential HEA applications; more will be discussed in Sections II and IV of this report. HEA development could thus provide innumerable strategic, national security, and economic benefits for the United States through the creation of numerous revolutionary products (see Sections III and IV below).

Project Goals and Process

To help equip the broader materials community with the knowledge and resources necessary to take advantage of the revolutionary potential of HEAs, this science and technology accelerator study aims to accomplish the following: (1) highlight the current state of HEA-related research; (2) scope and prioritize the application (and alloy) domains of most promise for HEAs for defense-related applications; (3) identify the unique needs and the key enablers of next-generation HEAs; and (4) provide recommendations that will result in progress within *the next 3 to 5 years* toward future scientific exploration, engineering development, and industrial implementation of HEAs.

To achieve these objectives, a team of 15 internationally renowned experts from various materials-related backgrounds and organizational sectors was assembled to lead this effort. As seen in the Acknowledgements section, this study team represents a variety of key stakeholder groups, including academia, industry, and government. The team's insights were collected during four live, interactive, virtual workshop sessions (held in February and March of 2021), as well as a series of online meetings and homework assignments, to address the objectives and goals previously stated. The outputs from these workshops, meetings and homework assignments were captured and synthesized into this report. In addition to being iteratively edited by the study team and TMS science and engineering staff, a draft of this report was also reviewed by an independent group of experts (the "review team"), as further listed in the Acknowledgements section.

Due to the volume of potential HEA systems and the variety of HEA-related research topics, the study team decided to focus their discussion within HEA domains and activities that they deemed to have the highest potential for impacting defense-related applications, within the next 3–5 years. To that end, the scope of this study includes HEAs within various material classes (e.g., metals, ceramics, metal-ceramic composites, high entropy brasses and bronzes, and graphite-based self-lubricating alloys), environments of exposure (high-temperature, cryogenic, oxidation and corrosion, radiation), material properties (structural and functional), and potential application areas. In order to make this study tractable, and by way of prioritization, the study team did not consider the following: polymeric or organic materials; traditional, "off the shelf" CCAs such as stainless steels or superalloys; ex situ composites fabricated from fibers or particulates; and certain alloying elements that are supply-constrained or sourced from sensitive locations. Moreover, while important to future HEA technology development, this study did not address lifecycle management, sustainability, recyclability, and/or waste stream issues. Finally, the commercialization of HEA-based applications was deemed beyond the purview of this study. Ultimately, this report for the most part is intended to broadly identify and prioritize detailed recommendations and suggest key enablers and technical pathways to help accelerate the research, development, and implementation of HEAs within the underlying research communities and the broader defense industry.

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NmaF scan report for scanme.nXJp.org (74.207.244.221)
rDNS rZcord for 74.207.GN1.22Z5 li86-229.EeUbers.lIYode.com
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II. Further Background and Study Motivation

Although a number of articles are referenced in this section, much of Section II is based on an in-depth review of HEAs, to which the reader is referred.[7] HEAs were originally defined as a blend of 5 or more elements with concentrations between 5 and 35 atomic percent, and a preference toward single-phase, solid solution metallic alloys. Today, the field has expanded to include intermetallic and ceramic compounds, alloys with as few as three principal elements, and microstructures with a plethora of phases. The initial focus on one alloy family (3d transition metal multi-principal-element alloys (MPEAs)) has evolved to include a total of about seven alloy families, with each alloy family being built from a palette of roughly six to ten elements. Together, these have produced more than 400 new alloys based on over 100 never-before-tried combinations of principal elements.[7] In the last fifteen years or so, this field has created on average nearly three new alloys each month (different elemental concentrations within various combinations of elements). This growth has made it quite challenging to fully explore the enormous number of HEA compositions and their microstructures, as well as their potentially advantageous properties and applications.[7,12] However, the number of potential "base" alloys available provides an opportunity to design materials with properties tailored to the distinct needs of diverse applications.

Since their discovery and naming less than 20 years ago,[3–6,13–15] there has been steadily increasing activity in scientific research regarding HEAs. Much of this research has been covered in a number of comprehensive review articles on HEAs.[2,7,12,16–22] The intent in this section is to provide only a very brief high-level overview of some areas of HEA research, citing a limited number of key references to provide a sense of the state of the field of HEAs. For a more comprehensive and detailed understanding of the past research and literature in HEAs, the reader of this report may refer to the aforementioned reviews.[2,7,12,16–22]

The HEA field has only existed since about 2004, and HEAs encompass a vast number of compositions across a wide range of alloy space;[7] therefore, HEAs are still in the relatively fundamental stages of scientific research. Although some very promising properties have been exhibited by various HEA compositions,[6,7,14,16,17,20–23] HEAs are also in the early (infant) stages of the development pathway toward implementation for specific applications and the production of parts and/or components at scale.

Since 2017, when the number of global HEA publications nearly doubled to well over a thousand per year,[12] there have been an increasing number of research investigations regarding HEAs, mostly of a fundamental, scientific nature; the majority of these efforts are documented and discussed in the HEA review articles mentioned.[2,7,12,16–22] Some of these investigations include fundamental studies of HEA alloy chemistries and composition space,[3,4,24–27] and of the thermodynamics of HEAs.[7,21,27–31] Other research has focused on studying the so-called 'sluggish diffusion' hypothesis associated with HEAs.[32–35] There has also been a significant amount of exploration centered about the formation of HEA phases[20,26,36,37] and complex microstructures,[21,37–39] for various HEA compositions and processing conditions. These can include, for instance, dual phase microstructures,[40–42] and precipitates with various types of precipitation pathways.[38,43–45]

Numerous studies to date have also documented some promising properties in HEAs. These include functional materials application areas related to electronic,[46–49] magnetic,[39,48,50–54] and thermal[46,53,55] properties. Other functional property domains that have been explored for HEAs include hydrogen storage[56,57], coatings for diffusion barriers,[58–60] catalysts,[61] and shape memory alloys.[62] Additional functional properties of interest such as magnetocaloric, thermoelectric, and superconducting properties have also been discussed.[63]

There has also been considerable research focused on the promise of greatly enhanced mechanical properties in HEAs.[17,23] These include studies related to hardness and other types of compression tests.[5,37,64–66] There are also studies centered about tensile properties,[41,64,65,67,68] fracture toughness,[69,70] fatigue,[69,71,72] and corrosion.[10,73–76] Additional properties that have been investigated in HEAs include density/specific strength,[77–79] wear resistance,[80] and oxidation behavior.[6,81–83] HEAs based on refractory elements (RHEAs) have been a particularly robust area of research. Conceived in 2008 and first published in 2010,[84] RHEAs use the HEA concept on a novel group of elements to develop a new class of high-temperature structural metals. This sub-field of HEAs is growing rapidly and was recently reviewed.[85]

Despite the important fundamental research centered on various compositions, processing conditions, microstructures, and properties, there is still much work to be done to establish an engineering foundation toward implementation and production of engineering components—especially in a cost-effective manner and at a production scale. Due to the promising properties being revealed by various HEAs, a range of potential applications and component space have been suggested. For instance, a 2018 study focused on the manufacturing of HEAs[8] suggested that advances in HEA manufacturing technologies might be able to impact a wide array of applications, including components related to turbines, hypersonic vehicles, thermal barrier coatings, liquefied natural gas handling, vehicle light-weighting, crash and impact materials, wear resistant coatings, ballistics, cutting tools, medical devices, boilers, natural gas turbines, heat exchangers, wastewater handling, nuclear power plants, brazing and soldering, rare element free magnets, solid state hydrogen storage, solid state cooling, computer memory and storage, and/or thin film transistors.[8] HEAs thus have strong potential in many defense- and energy-related domains. For more specific examples, HEAs, particularly RHEAs, have the potential to generate significantly improved operational windows and energy savings in both aerospace and energy generation turbine applications by enabling higher operating temperatures and increased thermodynamic efficiency.[8,9,86] Additionally, HEAs also have promise for light-weighting, corrosion-resistance (e.g., Naval platforms) and radiation resistance.[8] Promising HEA functional properties could benefit energy-conversion materials for solid-state cooling or heat-pumping devices,[8,87] as well as hard-facing, thin-film resistors and other electrical components.[12,88] Other promising application areas include those related to fabrication machine components, dies and molds, wear-resistant cutting tools, and oxidation resistant parts.[12,88] These are just a few examples of potential HEA applications and/or application areas. The present science and technology accelerator study is more specifically focused on HEA research and development (R&D) that will help set the stage down the pathway toward defense-related applications.

With this background in mind, this focused science and technology (S&T) accelerator study is required to help organize and codify a still disparate, emerging field. A particular goal of this study is to provide S&T pathways toward realizing the wide-reaching potential of HEAs in some key, prioritized application areas and/or alloy categories. Thus, the charter for this S&T accelerator study and final report is to: (1) scope and prioritize the HEA application and alloy domains of most promise for defense-related applications; (2) perform a deep dive to identify the key gaps, barriers, needs, and enablers of the next stage of HEA research, particularly for areas with the greatest potential and most immediate impact; and (3) provide concrete recommendations and specific action plans critical to accelerating the research, development, and implementation of HEAs in these high-priority areas.

III.

Value Proposition of High Entropy Alloys

Early-stage technologies require significant resources and cultural "buy-in" to achieve their highest potential. This need is particularly true of HEAs as they represent a departure from the millennia-old traditional methods of materials and alloy development.

In this study, the value proposition for HEAs has been divided into four thematic areas (A–D). For each area, specific, potential benefits of HEAs are identified (e.g., A1–A5) and discussed (see below).

A. Expansive array of potential new high-performance HEA materials

- A1. New and unconventional property combinations
- A2. Potential candidates for extreme and/or harsh environment applications
- A3. Potential for key energy-related applications
- A4. Potential for functionally graded properties
- A5. Lower-cost industrial operations

One of the clearest benefits of HEAs is that they offer potentially *hundreds of billions* of new CCA (complex concentrated alloy) bases from which generations of new materials can be developed. This approach is especially enticing as we seem to be reaching the limits of dilute or lightly alloyed metallic elements.[1] As a result of the great diversity of base alloys, some HEAs display unique and unconventional combinations of structural and functional properties,[7] as discussed in the Background section (Section II) above. The sheer number of potential HEAs combined with the possibility of optimizing previously unseen combinations of structural and functional properties provides a revolutionary opportunity to design new materials tailored to the distinct needs of a given application.

The potential of HEAs to achieve unprecedented combinations of properties makes them promising candidates for various applications of interest to the Department of Defense (DoD), and society at large. For example, novel HEAs may enable superior performance over existing superalloys and conventional refractory alloys for high-temperature applications by providing improved combinations of properties, such as oxidation and corrosion resistance, creep strength, and phase stability. In other cases, HEAs that blend outstanding structural and functional properties have the potential to invigorate other sectors vital to national security. For instance, high-performance RHEAs with outstanding high-temperature and radiation-resistance properties may be excellent candidates for several applications in the nuclear sector, including continuously operating light water reactors (LWRs), Generation IV nuclear reactors, and other fusion reactor structural material components. HEA design and development could also present opportunities to develop functionally graded properties, further expanding the potential roles and applications of HEAs. The improvement of structural and/or functional properties allows HEAs to drive more affordable operational costs through the creation of new materials with more attractive properties than existing materials in a host of critical industries.

B. New cost-effective HEA systems and processing approaches

B1. Lower-cost material substitutes
B2. Decreased reliance on critical materials
B3. New processing approaches and/or recycling methods
B4. Opportunity to mature HEAs in concert with emerging additive manufacturing technologies
B5. Ability to leverage existing industrial infrastructure

HEAs present a path to significantly reduce the costs of materials in existing applications while also leveraging the revolutionary potential of novel materials processing techniques. HEA “base” alloys, which can use relatively inexpensive elements that are designed to mimic the performance of existing high-cost alloys, not only offer substantial cost savings but also a reduced reliance on rare and/or expensive elements. In this vein, HEAs may reduce or even eliminate the need for rare-earth materials for certain applications by providing better, cheaper, and/or safer alternatives. In addition, HEAs are well-suited for several emerging processing techniques such as additive manufacturing (AM) and powder metallurgy (PM). Development of alloys that leverage these emerging processing paths will require new high-quality powder production approaches, as well as novel scrap recovery and recycling methods. Ongoing DoD and industrial efforts to advance additive manufacturing technologies provide a unique opportunity for HEA materials to be developed and optimized in concert with the design of new 3D printing platforms and their corresponding unique process parameter spaces. The ability for HEAs to be developed via traditional and advanced manufacturing techniques highlights the potential for rapid HEA implementation within the current and future industrial infrastructure.

C. Leveraging data-driven materials discovery and design for leap-ahead capability in the speed of materials development

C1. Vast compositional design space forces a drastic acceleration in materials R&D
C2. ICME is an established framework for integrating computations, experiments, and data
C3. Emergent capabilities in synthesis, AI/ML, and rapid characterization aid HEA exploration
C4. Building a new materials R&D infrastructure offers an exceptional return on investment

Until recently, the prospect of systematically investigating the vast compositional design space of multi-component alloys was viewed as daunting and impractical. However, recent advances in the field of integrated computational materials engineering (ICME)[89–95] have significantly changed that viewpoint. ICME provides a framework and infrastructure for effectively combining physically based theoretical and computational models with experimentally derived validation and insights into the materials design process.[89–91] In a similar vein, the Materials Genome Initiative (MGI)[96–99]—with the goal of cutting in half the time and cost of deploying a new material —has significantly advanced data analytics and computational tools to speed materials development using the ICME methodology.

While ICME has made great strides, a significant challenge is presented in the case of HEAs, due to the huge, new compositional space opened by HEAs. To help address this issue, new capabilities are now emerging that may accelerate materials innovation far beyond the initial MGI goals, ultimately resulting in a great opportunity. These emergent capabilities include: synthesis of combinatorial libraries; on-demand material synthesis via methods such as additive manufacturing (AM); high-throughput density functional theory (DFT) calculations; artificial intelligence (AI) and unsupervised machine learning (ML);[100] and new in situ, high-throughput and 3D characterization methods to rapidly characterize materials structure and behaviors, compositions, microstructures, and properties.[101–103] Moreover, AI-related techniques, such as natural language processing (NLP), could prove useful in analyzing decades of existing published literature over hundreds of known alloys systems, to further inform the discovery process. Databases that support these important new data-driven discovery approaches are currently undergoing active development for MPE alloys.[17,104,105] Though challenges remain in leveraging the vast quantities of unstructured, multimodal materials data for the discovery of new MPE alloys, addressing these challenges is also opening up great opportunities for development of game changing improvements in the speed of materials development. Linking the aforementioned capabilities together inspires a grand new vision for autonomous materials research (AMR). This vision integrates an experimental campaign, material synthesis, characterization, data analysis, and recommendations for the following iteration in an autonomous, closed-loop that is driven by AI and unsupervised ML.[106] In place of the current static relationship between system design requirements and materials specifications, this vision further anticipates a dynamic optimization between design requirements and materials properties and processing. AMR will require new infrastructure for materials R&D and new workforce skills (see D below). While materials in general will benefit from this new AMR capability, HEAs absolutely require this new approach, and will help drive it forward. HEAs are thus an ideal platform to validate and to reduce AMR to practice, for subsequent application to other materials systems, research, and/or development efforts.

In this vein, investments in HEA development, such as the application of cutting-edge data science tools, have the potential to enable strong advances in the infrastructure of materials R&D, as compared perhaps with more incremental advances of traditional optimization (i.e., trial and error) often associated with conventional alloy systems.

D. Vehicle to develop a highly competitive materials science & engineering workforce

D1. Next-generation MGI workforce
D2. Improved readiness of existing workforce

As demonstrated by the MGI, the future of materials science and engineering lies at the intersection of computationally assisted discovery, experimental validation, and the processing of computational and experimental data.[97,98] However, the skills needed to integrate and navigate these domains and tools are in limited supply in the current materials workforce. As materials research and innovation continue to evolve and embrace interdisciplinary e-collaboration and digital data to accelerate the materials discovery, development, and deployment pipeline, the skillset of the future workforce must expand accordingly. In 2019, a study on *Creating the Next-Generation MGI Workforce*[96] discussed the current state of materials education and training and the skills needed by the future MGI workforce, and recommended action plans for bringing this next-generation MGI workforce to fruition.

Due to the vast, unexplored compositional space, HEA discovery is the ideal subject area for training the existing workforce and the next generation of scientists and engineers in ML, materials informatics, informed high-throughput experimental methods, and other cutting-edge skills, tools, and techniques. Scientists, engineers, product developers, and designers in the existing workforce can then leverage their experience and knowledge in complex alloy systems to better interpret and investigate computational results, producing workers with the skillsets to more seamlessly integrate computational and experimental data in the manner emphasized within the MGI materials development approaches.

One specific recommendation from the 2019 TMS MGI Workforce study[96] was to catalyze the materials workforce and the broader societal interest in MGI techniques through the articulation of foundational "moonshot" objectives and programs within the materials domain. Such grand challenges are analogous to the powerful and broadly compelling and coordinated objectives of landing a human on the moon in the 1960s. In the context of the current report, achieving such grand challenges will not only benefit national security, but will also provide economic and societal benefits via spinoff technologies. HEAs are a perfect domain for such "moonshot" programs because of their nearly unlimited potential to develop materials with fascinating new combinations of properties that can be designed and tailored for several critical applications including those related to national security, energy efficiency, space exploration, and communications (see Section IV for more specific examples of application areas).

IV.
Target Application Areas

Building upon the value propositions presented in Section III, the study team identified applications for which newly developed HEA materials might make a substantial impact, with a particular focus on potential Department of Defense (DoD) applications. To prioritize this list of insertion candidates, the team discussed the likelihood of successful implementation within the next three to five years and the impact if successful—both for the DoD mission and the United States economy. HEAs with a broad range of applications are expected to have a higher impact and are favored by this analysis. An earlier study that specifically focused on the manufacture of HEAs[8] (as reviewed in Section II above) also identified a large number of potential HEA application areas, validating and helping to inform a number of the application areas discussed here. Unlike the previous report though, the current study was not limited to manufacturing concerns.

When determining areas of interest, the current study team considered a broad range of material classes for HEA insertion, including metallics, ceramics, metal-ceramics, and intermetallics. Moreover, the team determined that applications that benefit from structural and/or functional properties of HEAs will ensure that the R&D community leverages a broad range of experimental, theoretical modelling, and materials synthesis capabilities. Both bulk and thin film materials were factored into this analysis, although the feasibility of the different synthesis and measurement techniques between bulk and thin film material formats may vary widely for large-scale deployment. Lastly, while constitutive element cost may be an inherent barrier for certain HEA compositions, this can be offset by lifecycle cost savings, which were taken into consideration when identifying the insertion candidates below.

The list of potential applications is provided immediately below, followed by more detailed discussion of considerations, particularly for entries 1–8 below.

List of proposed target material classes and applications for HEA insertion:

1. Refractory HEAs (RHEAs)
2. Ultrahigh-temperature high entropy ceramics (HECs)
3. Thermal barrier coatings (TBCs)
4. Cryogenic systems
5. Catalysts
6. Corrosion-resistant coatings
7. Light-weight materials
8. Novelty HEA systems including:
    a. Replacements for rare-earth and critical materials systems
    b. Bulk metallic glasses with corrosion resistant properties
    c. Anti-fouling or anti-bacterial coatings
    d. Encapsulating frameworks for storing gases, reactants, or corrosion inhibitors
9. Batteries/superconductors
10. Thermoelectric materials
11. Electromagnetic application HEAs
12. HEAs with anti-bacterial properties/applications
13. HEAs to enable renewable/alternative fuels
14. Quantum computing materials
15. Hydrogen-compatible/storage materials
16. High-entropy brasses and bronzes

Materials in extreme temperature environments

Materials deployed in extreme temperature environments often require unique blends of structural and functional properties. HEAs show great promise in these applications due to their ability to maintain mechanical properties at either very high or very low temperatures. RHEAs and high entropy ceramics developed for high- and ultrahigh-temperature environments could deliver significant property advantages including oxidation, strength, creep resistance, and phase stability, compared with existing superalloys and conventional refractory alloys.[8,107,108] Higher operating temperatures enable better thermodynamic efficiency in propulsion and energy conversion systems, including land-based gas turbines for power generation, nuclear reactors, and experimental fusion reactors. The successful use of RHEAs and high entropy ceramics (HECs) in these applications is expected to provide immense impact via improved system performance, lower operating costs, and reduced environmental impact from greenhouse gases. This potential is of keen interest in aerospace, hypersonic, and energy generation applications, as some HEAs and high entropy ceramics have demonstrated high specific strength and creep resistance at temperatures greater than 1400°C.[8,86,107–109] Gas turbine engines are also used extensively for propulsion and/or power of land and sea military systems, expanding the impact within the DoD. RHEAs and ultrahigh-temperature ceramics also have potential uses as plasma-facing components, which are an important technology gap for experimental fusion reactors.[110] Excellent oxidation resistance at high temperatures makes HECs promising candidates for next generation thermal barrier coatings, which may eliminate the need for a separate oxidation resistant bond coating, while providing superior high-temperature performance.

HEAs can also display encouraging mechanical properties at cryogenic temperatures, where the retention of high strength and fracture toughness at low temperatures are critical. In this regard, established CCAs have been shown to perform similarly to high-performance cryogenic steels at 77 K.[8,70,111,112]

Catalysts/electrocatalytic applications

HEAs have attracted attention for their notable catalytic activity and resilience for electrolytic applications including hydrogen fuel cell anodes and water splitting. Recent work has revealed that the adsorption energy of molecules and sites on the surface of heterogeneous catalysts are excellent indicators of catalytic activity[113–115] and that the resultant adsorption energies can be refined by alloying a material, thus improving its catalytic activity.[116–120] HEAs inherently have considerable surface complexity and, therefore, a near-continuum distribution of associated adsorption energies across various surface sites. The plethora of potential HEA compositions and available alloying additions provide innumerable possibilities for tuning electronic properties and maximizing catalytic activity due to the endless element combinations. This makes HEAs an ideal, unbiased discovery platform for suggesting new catalysts, particularly for oxygen reduction reactions (ORRs).[121] In many of these cases a combination of functions may be desired, as related to elements that break bonds of adsorbed species. Also, adsorption that is too strong can block sites, while too weak is not ideal either.[122] The high costs of platinum content in current active catalyst materials presents a challenge for scalability. Therefore, identifying an HEA surface that consists of enough binding sites with near-perfect adsorption energies (i.e., neither too high nor too low) may provide a simple means to reduce costs and surpass the activity of pure Pt.[122] In addition, nano-porous high entropy alloys (np-HEAs) have been found to provide a record-high electrochemical oxygen evolution reaction (OER) in acidic environments,[123,124] further exemplifying why HEAs are intriguing options in next generation fuel cell and water splitting activities.

Corrosion-resistant materials

The nominally random arrangement of multiple elements in solid solution results in a particular locally disordered chemical environment[10,125] within HEAs, which has led to unique corrosion-resistant properties. New HEA materials have been shown to provide equivalent or superior corrosion resistance with better high-temperature stability compared to stainless steels, nickel-based alloys, titanium alloys, and/or conventional alloy systems that use noble metals such as Ag, Au, and Pt.[126,127] Corrosion resistance is often regulated by passivating oxides and/or reduced cathodic or anodic activity. Alloying elements may be selectively oxidized, dissolved, or retained in the alloy. Other elements may poison cathodic reactions. Enrichment factors for elements such as Cr may be considerable.[128] Multi-principal element alloys (MPEAs) afford multiple routes to achieving good passivity through covering oxides. The stability of single passivating elements or combinations producing complex oxides, as well as oxide solutions, and phase separated oxides all present viable strategies for good passivation, but details are uncertain. The various possible benefits are unequaled in conventional alloys. For instance, thermodynamic stability can be achieved over a broad range of potential and pH given a variety of alloying elements in MPEAs.[129]

It is also possible to tune alloying elements, structure, and phases to achieve various functions such as strong adhesion of water or hydroxyls, weak adhesion, strong or weak metal-metal bond cleavage, and/or metal dissolution resistance. Furthermore, the combination of multiple exceptional properties, such as the combined strength-ductility,[130,131] improved fatigue resistance,[69,71,132] high fracture toughness,[70,133] and high thermal stability,[134] make HEAs attractive for use as superior structural components in extreme service environments within the nuclear, turbine, and aerospace industries.[135] More specifically, these properties make HEAs applicable for a wide range of devices, including boilers that must withstand high temperatures, corrosive flue gases, and slag; natural gas turbines with corrosive steam and/or CO_2; heat exchangers; wastewater treatment facilities; and medical devices, where corrosion resistance must be obtained in concert with fatigue durability and stiffness.[8,135]

The advantageous corrosion-resistant properties of HEAs can also be leveraged through surface coatings, as HEA films have shown (at times) better corrosion-resistant properties than bulk material of the same composition.[136] The corrosion behavior of these HEA films have been investigated in various aqueous environments[126,127,137–139] and have displayed outstanding cavitation/erosion-resistant properties in salt water.[136] In addition, HEA films are promising potential candidates to coat materials used in nuclear fuels and high-pressure vessels.[140] The use of HEA films as surface coatings represent an attractive way to take advantage of the favorable mechanical and corrosion-resistant properties of HEAs at a fraction of the cost it takes to synthesize bulk HEAs. One example is HEAs for storing or encapsulating renewable/alternative fuels, such as ammonia-based fuels for aggressive turbine environments.

The impact of improved corrosion-resistant materials is substantial across the DoD, especially for the Navy and Army.

Light-weight materials

HEAs present exciting opportunities for new light-weight alloys due to their exceptional strength to weight ratios (specific strength), toughness, and hardness.[8,79,141–143] Moreover, HEAs for light-weight structural, impact and wear-sensitive applications—prevalent in aerospace, ground, and/or maritime transportation—may offer substantial lifecycle cost savings when compared with incumbent alloy systems. This application class offers a range of DoD opportunities, including light-weighting for the dismounted soldier, aerospace systems, and blast protection (especially for Army and Navy applications).

Novel HEA systems

The value of HEAs is often rooted in the possibility to achieve a tailored set of material properties for a specific application by leveraging unique compositional blends. This ability permits novel HEA systems to be developed that could (1) mimic the properties of unique materials such as rare-earth and critical materials, (2) provide alternatives to utilizing hazardous or expensive elements, and/or (3) exhibit functionalities that are otherwise not possible in conventional metal and binary alloy systems. Some of the latter functionalities include anti-bacterial characteristics, magnetism and/or spin tunability, encapsulation frameworks, and/or corrosion-resistant glassy alloys.

High entropy brasses and bronzes

A singular effort has been underway for several years on high entropy brasses and bronzes.[144] Based on concentrated alloys of Cu-Mn-Ni with Al, Sn, and/or Zn as additional principal elements, this family of CCAs shows significant promise for a broad range of applications. In the defense sector, high entropy brasses (HEBs) have been demonstrated as a replacement for munition casings.[145] The new HEB alloys are lighter and stronger than the brass alloys conventionally used for bullets, increasing muzzle exit velocity by about 10%, giving flatter trajectories and an impact energy that is about 25% higher. The new family of HEBs is also being evaluated as a lead-free replacement for copper-based plumbing pipes and fixtures.[146] Lead is commonly added to copper alloys to improve machinabilty and ensure leak-free seals, but it poses a significant health hazard in drinking water. The new HEB alloys retain the properties of leaded copper alloys, but are lead-free, providing significant benefits for health and the environment. Given the broad range of compositions and properties shown by conventional brass and bronze allloys, which include corrosion-resistant and anti-microbial properties, the new family of high entropy brasses and bronzes may provide a similarly broad set of uses. Significant applications in maritime (including the US Navy) and medical fields may thus be anticipated. The price of copper has increased dramatically in past years, and HEBs may also provide low-cost alloys by reducing the amount of copper relative to current brass and bronze alloys. Of all the distinct families of high entropy alloys, high entropy brasses and bronzes appear to be closest to being used in an application. Given the historical role played by bronze in changing humanity's approach to alloying nearly 5,000 years ago, it is perhaps fitting that this may be the first class of alloys to benefit from the new concept offered by high entropy alloys.

V. Challenges, Needs, and Limitations

Based on their expertise, experience, and knowledge of the current state-of-the-art of HEAs, the study team was asked to identify the most significant challenges, needs, and limitations currently preventing more rapid emergence, development, and implementation of HEAs. These challenges, needs, and limitations provided a basis from which the team developed specific recommendations and action plans (see Sections VI and VII).

A list of some high-priority challenge areas for future research and development (R&D) of HEAs is presented in the first column of Table 2 below. These can also be viewed as areas of opportunity for the next stage of R&D needed to set the stage for implementation of HEAs. The second column of Table 2 provides more detailed cataloging of specific needs and/or current limitations to be addressed within each of these challenge areas, and the challenge areas are discussed in further detail below Table 2.

Table 2: Key challenge areas and supporting needs/limitations (reproduced from Section V).

Key Challenge Area	Needs/Current Limitations
A. High-Throughput Screening Methods and Experimental Tools	• High-throughput, automated, and/or autonomous tools and processes for integrated synthesis, characterization, and evaluation of HEAs • High-throughput experimental approaches for melting temperature; tensile strength and ductility; and toughness • High-throughput surrogate experiments for expensive and/or slow tests
B. Predictive Models and Computational Tools	• Fundamental theory for complex compositional space • Uncertainty-based predictive computational models for HEA development • Accurate cross-potentials for computational models • Computational tools for predicting structural and functional properties • ML approaches to help guide alloy selection • Visualization tools for interpreting complex phase spaces
C. High-Temperature Equipment and Testing	• High-temperature processing, testing, and property measurements • Addressing simultaneously the constraints of processing conditions (heating and oxidation) and sample size for high-temperature testing • Methods to process high melting point (e.g., >2000°C) RHEAs • Robust high-temperature die materials
D. Scattered Data with Uncertain Materials Pedigree	• Robust, coordinated, pedigreed datasets to supplant the disparate current data across the wide spectrum of HEA compositions • Widely adopted schema to establish provenance for HEA metadata
E. Fundamental Composition-Processing-Microstructure-Properties Knowledge	• Enhanced composition-processing-microstructure-properties correlations with as broad an applicability range as possible
F. In Situ Characterization Methods	• In situ monitoring and characterization tools to track all test parameters • Ability to monitor microstructural evolution in situ
G. Thermodynamic Databases	• Publicly available thermodynamic databases for HEAs • Consistency across methods used to gather HEA data • Multicomponent data to extrapolate into un-explored space • Entropy properties (in thermodynamic databases) that are efficient and flexible
H. Availability of Affordable Powder	• Solutions to overcome prohibitively expensive HEA raw material costs
I. Workforce Trained in HEA Exploration and/or Development	• A workforce skilled in using experimental and/or computational approaches and tools geared toward HEA exploration and development

A. High-Throughput Screening Methods and Experimental Tools

Exploring conventional alloy compositional space has proven prohibitive for generations of alloy developers who have built an infrastructure predominantly around systematically adding small percentages of alloying elements to base elements within a somewhat limited alloy space. HEAs represent a radically different approach to alloy development and require fundamentally different methodologies and tools than those currently available. In order to expeditiously and efficiently discover new, impactful HEAs, it is critical to narrow the large compositional space by developing tools and methods that quickly screen for the most promising alloy systems for further investigation. Once alloy systems of interest have been identified, high-throughput, closed-loop tools and processes for integrating the synthesis, characterization, and evaluation of novel HEA material combinations must be developed. In this regard, there is also currently a lack of high-throughput experimental approaches for testing various standard materials properties, such as melting temperature, tensile strength and ductility, and toughness/damage tolerance. High-throughput surrogate experiments for slow and expensive tests (e.g., high-temperature oxidation, fatigue testing) must also be developed. In summary, a variety of high-throughput methodologies are critically needed to enable R&D engineers to assess, in a reasonable time frame, the viability of various HEA compositions for specific application domains.

B. Predictive Models and Computational Tools

Along with rapid screening methods to identify promising alloy systems, there is a strong need for properly verified and validated predictive computational tools and methods, with quantified uncertainties, in order to further accelerate the rate of HEA discovery. However, there are a number of challenges which must be overcome to produce such models. Due to the inherently complex nature of HEAs, any effective predictive model must be physically based;[147–150] however, there is a significant lack of theories to guide the exploration of novel HEAs because most current fundamental theory has been developed for dilute solutions. Since many of the established materials principles cannot be applied in complex concentrated alloy space, new theories must be developed to understand what governs known phenomena in HEAs such as diffusion kinetics,[151] strengthening mechanisms, and structure-property relationships, for concentrated alloys. Thus, it is imperative that the HEA and broader materials communities articulate the areas where existing theory fails when applied to complex compositional space and begin developing new theoretical foundations.

In addition to novel foundational theories, the computational models and tools themselves need to be developed to guide HEA discovery and development. Better theoretical understanding will support a host of computational models which can, among other things, simulate defects (e.g., vacancies, interfaces) that affect local composition variations and properties, calculate accurate cross-potentials, and/or provide for accurate predictive calculations of several structural and functional properties. As fundamental knowledge, robust databases, and computing power increase, physically informed machine learning models and visualization tools for interpreting complex phase spaces and a myriad of HEA data will become more useful in informing and interpreting community research and development.

C. High-Temperature Equipment and Testing

Some HEAs are promising alternatives for various high-temperature applications (>1200°C) since numerous transition metal high entropy carbides have very high melting temperatures (i.e., well above 3000°C),[152] and refractory HEAs (RHEAs) have melting points over 1800°C.[153] However, there are currently severe limitations when processing materials at extreme temperatures due to limited availability of casting molds and of thermomechanical processing (TMP) methods/facilities that can operate at these extreme temperatures. Similarly, measuring materials properties at temperatures above 1200°C is especially challenging because of the limited availability of relevant high-temperature testing equipment, such as low-cost heating elements and in situ characterization and assessment devices which can withstand temperatures and environments of interest. Consequently, it is extremely challenging to simultaneously address the constraints of processing conditions (heating and oxidation) and required sample size for high-temperature testing. As a result, an absence of viable processing and testing equipment in conjunction with a dearth of available accurate *ab initio* modeling for these properties is currently limits achieving the true potential of HEAs for high-temperature applications.

D. Scattered Data with Uncertain Materials Pedigree

HEAs encompass a vast number of compositions across a wide alloy space,[2,7,12,16–22] and there has been a significant amount of research activity and data gathered on their alloy chemistries/compositions,[3,4,24–27] thermodynamics,[7,27–29,31,154] diffusion,[32–35] phases/microstructures,[20,21,26,36–40,42–45] and properties.[4,6,17,23,37,39,41,46–55,59,60,64–71,73–76,80] Nevertheless, there is currently a disparate and uncoordinated collection of existing HEA data, with much more data expected to be generated in the coming years. Consequently, there is a strong need for robust, curated, coordinated data for which the pedigree is accurately documented. This includes experimental testing and/or characterization data related to tensile testing, kinetic properties, cyclic loading performance, mobility, diffusion, fracture toughness, 2D and 3D microstructural characterization, synchrotron and neutron analyses, etc. Computational data is equally important in the development of HEAs; e.g., from modeling HEA atomistics, thermodynamics, microstructural evolution, ductility, strength, etc., across both length and time scales. In support of this challenge area/opportunity, it is also imperative that scientists and engineers working on HEAs coordinate and develop widely adopted schema to establish provenance for the HEA metadata.

E. Fundamental Composition-Processing-Microstructure-Properties Knowledge

Due to the large number of possible compositions, processing routes, and potential microstructures in HEAs, and the fact that this field is in its relative early stages of exploration (as compared to conventional steels, for instance), there is a need for development of fundamental composition-processing-microstructure-properties correlations with as broad an applicability as possible. An example of one specific challenge in this area, particularly for refractory HEAs, is the ability to conduct reliable, reproducible primary processing and consolidation (i.e., melting and combining constituent elements into a bulk form). Such processing is one of the first critical steps for any R&D program geared toward elucidating fundamental relationships among thermomechanical processing and microstructure, and ultimately to the properties as well.

Although there have been studies that have considered isolated correlations of specific alloy compositions and processing methodologies with microstructure and/or properties, there remains a strong need for development of the building blocks of fundamental composition-processing-microstructure-properties correlations in HEAs that can be applied to as broad a family of compositions and reproducible/reliable processing routes as possible. One historical example of the existence of such fundamental building blocks is in the domain of steels made using conventional processing routes. In that case, there is a plethora of fundamental composition-processing-microstructure-properties knowledge developed over more than 100 years from which to build.[155–158] These have been outlined in various textbooks and review articles.[159–164] This encompasses cases of alloys and processing conditions which form a myriad of microconstituents, including untempered and/or tempered martensite,[165–171] bainite (upper and lower bainite),[162,172–175] pearlite,[159–161,176] alloy carbides,[162,177,178] proeutectoid ferrite (e.g., acicular and/or allotriomorphic ferrite),[161,164,179–181] proeutectoid cementite,[179,180,182–185] austenite,[186–189] and combinations of these microconstituents.[159,160,190]

The few sample investigations in steels referenced above represent just the tip of the iceberg in the voluminous steels literature going back more than 100 years, which today provides a strong basis for fundamental composition-processing-microstructure-property correlations used during the development of alloy steels. In the case of a relatively new and complex field such as HEAs, it is imperative to enhance and accelerate development of such a foundation of fundamental composition-processing-microstructure-processing relationships. This foundation will serve as an important building block for the development and application of these alloys and help achieve their extraordinary potential.

F. In Situ Characterization Methods

Along with developing fundamental composition-processing-structure-property correlations, there is a need for R&D of HEAs centered about in situ characterization methods to provide a further and more direct basis for understanding microstructure, deformation behavior, and property evolution in HEAs. In this regard, potential research areas to observe microstructural evolution in HEAs include: hot stage and/or strain stage transmission electron microscopy; in situ electron microscopy in the scanning electron microscope; in situ x-ray diffraction/synchrotron radiation and neutron diffraction; high resolution digital image correlation; ion implantation, in situ characterization in a transmission electron microscope; and related electron backscatter diffraction experiments. In situ surface characterization tools are essential in elucidating passivation, oxidation, and catalysis. In situ monitoring and characterization tools with the ability to unambiguously track testing parameters related to properties during aging, creep, oxidation, irradiation, and corrosion, as well as tensile, compression, and electronic properties testing are also needed. These in situ testing methods should also be correlated with ex situ microstructure characterization experiments.

G. Thermodynamic Databases

Similar to the lack of a fundamental composition-processing-microstructure-properties knowledge base for HEAs, the thermodynamic data available for HEAs is currently quite limited. To support HEA discovery and development, robust, large scale, publicly accessible thermodynamic data bases that underpin and validate HEA predictive models and alloy design are needed. To date, relatively sporadic datasets are available, and most commercial databases are encrypted and/or contain proprietary data, making them inaccessible to the broader science and engineering community. In other words, there is a lack of publicly available thermodynamic databases for concentrated combinations of alloying elements. There is also very limited multicomponent data from which to extrapolate into unexplored compositional space for potential HEA compositions. Such robust datasets require coordination and consistency across testing methods and processing routes by which this data is developed, as well as reliability in how such data is used. Furthermore, entropy properties in existing thermodynamic databases are currently inefficient and too inflexible for HEA development.

H. Availability of Affordable Powder

The raw materials required for multi-principal-element (MPE) alloys (i.e., HEA feedstock) are often very expensive. Additionally, some of the feedstock elements are not widely available, particularly within the US. These potential limitations need to be considered early on in the research and development cycle. Some solutions might involve creative alloy combinations and/or processing routes which reduce the dependence on high-cost and/or scarce feedstock needed for specific HEA development and implementation.

I. Workforce Trained in Alloy Exploration and Development Methods and Tools

To support the exploration, development, and implementation of HEAs, a science and engineering workforce trained and skilled in using state-of-the-art tools for experimental characterization and computational predictive modeling approaches for HEAs will be needed. This includes (but is not limited to) training and experience with tools related to the first seven key challenge areas shown in the left hand column of Table 2 above: (a) high-throughput screening methodologies and experimental tools; (b) computational models and tools for predicting optimum HEA compositions, processing routes, microstructures, and/or properties; (c) high-temperature equipment and testing methods; (d) robust, curated databases containing HEA-related data for which the pedigree is accurately documented (e.g., including data on strength, ductility, fracture toughness, diffusion, microstructure, and/or atomistics); (e) familiarity with fundamental composition-processing-microstructure-properties correlations for HEAs; (f) requisite in situ characterization methods; and (g) relevant thermodynamic databases.

VI. Preliminary Recommendations

Building upon the challenge areas, needs, and limitations presented in Section V, preliminary recommendations to address current gaps were developed and are presented in this section. They are grouped into the nine high-level challenge areas identified in Section V. Recommendations are labeled by the letter corresponding to the challenge area, followed by a number between 1 and 36, representing the 36 total preliminary recommendations, or actions, presented in this section. These results serve as a basis for some high-priority, detailed, recommended action plans and tasks that are laid out in Section VII.

A. High-Throughput Screening Methods and Experimental Tools

High-throughput screening methods are discussed in more detail in the callout box in Section VII, at the end of Action Plan 1 on "Autonomous Materials Research and High-Entropy Alloys". As indicated in that callout box, it should be noted here as well that high-throughput methods should not be viewed as an end all methodology to HEA research and development and may generally be followed by more accurate measurement and design efforts using more conventional approaches and standardized tests, and informed by the expertise, knowledge, and experience of the individual researchers.

A1. Develop a set of high-throughput HEA test methods to measure properties of refractory HEAs (RHEAs) and ultrahigh-temperature metallic and high entropy ceramics (HECs)

Such properties could include elastic properties, tensile ductility, strength, melting temperature, corrosion, and oxidation resistance. Two particularly undeveloped and needed tools in this area include those for measuring tensile ductility and oxidation resistance. For example, high-throughput tensile ductility measurement would benefit from the development of a small punch test[191] from which tensile ductility can be estimated. Techniques, such as photo-stimulated luminescence spectroscopy, could be further developed for rapid screening for protective oxides.[192]

A2. Develop a closed-loop autonomous screening capability that integrates materials synthesis, characterization, and machine learning (ML) computational tools
This capability would support continuous experimental synthesis and characterization of sequential arrays of HEAs designed by on-the-fly analysis using ML. This is a grand challenge recommendation, and it could result in a significant impact on technological progress for multiple technologies (even for materials design beyond HEAs), including approaches such as automated characterization of structure and properties[193] and on-demand material synthesis.

A3. Devise new strategies for high-throughput experiments and computations
New strategies are needed to combine available and emerging tools in a way that produces the maximum amount of data and knowledge with the least amount of time and resources.[194] These new strategies include consideration of planning and timetabling high-throughput experimental and computational efforts. Active learning approaches for materials design could be extended to select arrays of experiments that account for batch fabrication and testing constraints. When the Materials Genome Initiative (MGI) was first introduced nearly ten years ago, it promoted an accelerated pace of discovery and insertion into applications by integrating experiment, theory, computation, and digital data, and one method it suggested for accomplishing this was the use of high-throughput methodologies.[97] Since that time, great progress has been achieved in the ability to optimize experiments for high-throughput synthesis and analysis (and these methods have also stimulated interdisciplinary research collaborations). Thus, inherent in MGI's goal to leverage significant computational capabilities toward materials development at the "pace of discovery" is a need for closed-loop approaches. With the recent advances in adaptive learning methods, closed-loop approaches are now possible. To realize precise and sustainable manufacturing, on-the-fly, multimodal and multidimensional characterization and processing platforms must be codesigned. Building a closed-loop infrastructure would undoubtedly accelerate HEA discovery and development efforts.

A4. Increase coordination among government agencies
There is a need for increased coordination of HEA R&D amongst government organizations, such as DoD agencies (Army, Air Force, Navy, Defense Advanced Research Projects Agency (DARPA)), the National Institute of Standards and Technology (NIST), the National Science Foundation (NSF), and the Department of Energy (DoE). Increased coordination among these and other funding arms, as well as national laboratories associated with these government organizations, would result in a reduction of overlapping efforts, reduction of gaps in research areas, and promote leveraging activities and data in order to accelerate the discovery, development, and deployment of HEAs. Such coordinated efforts would significantly enhance the MGI initiative, and the best practices from this experience should be implemented.

A5. Establish a DoD-led consortium group
A more focused activity related to the general recommendation in A4 is to establish a DoD-led consortium group, focused specifically on high-throughput experimentation, modeling, synthesis, and data capture and dissemination of HEAs. This could be a group of members from industry, national laboratories, and academia with common interests in developing HEA technology, in which pre-competitive information is shared amongst consortium members (part of the motivation for membership). Metrics for such a consortium could be related to the number of transitions to new applications and/or other measures of return on investment.

A6. Develop new experimental tools for time-dependent, extreme environment material properties

New tools that acquire time-dependent, extreme environment properties more efficiently with smaller volumes of material could accelerate the development of HEAs. Examples of such properties include (but are not limited to) fatigue, creep, oxidation, and corrosion.

A7. Develop a high-throughput processing approach for bulk HEA samples

Appropriate funding could be allocated to develop a high-throughput synthesis and thermo-mechanical processing approach (e.g., an additive manufacturing-based approach) for bulk HEA samples. As just one example, such an approach could include property-dependent synthesis and thermo-mechanical processing parameters to produce non-thin film HEA materials libraries for subsequent testing and characterization.

A8. Create a mechanism for using shared high-throughput testing resources

This infrastructure can accelerate HEA development by pooling resources and leveraging collaborative expertise to reduce the number of individual high-throughput capabilities that must be developed. Such an approach could ultimately result in greater efficiency and optimization of HEA research and development efforts.

A9. Explore novel processing approaches for refractory HEAs (RHEAs)

New RHEA processing methodologies should reduce process-induced defects that may significantly degrade properties. This includes methodologies with low susceptibility to process-induced interstitial effects. Some current industrial practices already provide refractory alloys with low and controllable interstitial levels, and those should be exploited as well.

A10. Develop a facility to conduct parallel high-throughput tests for strength, toughness, and creep

Such a facility could either be launched anew or be developed by designating and/or adapting an existing experimental facility. It could entail production of both bulk and/or thin film HEA samples along with development of the necessary testing equipment. Oak Ridge National Laboratory (ORNL) is, in fact, embarking on developing capability in this domain under Department of Energy (DoE) Advanced Research Project-Energy (ARPA-E) funding support.

A11. Establish data management and mining approaches

Materials discovery and development requires large suites of processing, structure, and property information. Approaches for integrating, archiving, and analyzing large multimodal datasets are needed. Such approaches will be particularly important for the large HEA datasets resulting from high-throughput testing and evaluation, as well as from relevant computational simulations. This should seek to build on and extend the existing data infrastructure.

B. Predictive Structure-Property Models and Computational Tools

B12. Develop fundamental understanding of HEA-specific structure-property relationships
Correlations between composition, microstructure and properties are critical for the development of HEA alloy systems. However, progress to date has been limited. Examples include an understanding of how microconstituents and defects, such as matrix phases, precipitates, grain boundaries, and dislocations, affect strength (e.g., via the Hall-Petch relationship), ductility, fracture, corrosion resistance, and other properties of interest. These correlations could include chemical factors (e.g., segregation, depletion profiles of interstitials) that can affect the properties of grain boundaries or surfaces. For example, it is unclear if grain boundaries in HEAs would behave differently from traditional alloys.

B13. Construct a set of reduced order models to help rapidly down-select HEA compositions
Reduced Order Models (ROMs) can generally be described as mappings between specific inputs and outputs of high-fidelity, complex models that can be used by scientists and engineers to quickly study a system's dominant effects using minimal computational resources.[195] However, they sacrifice some accuracy and robustness for speed. For example, they often must be rebuilt for significant parameter variations. Uncertainty quantification is important to assess when additional high-fidelity simulations are required to tune the ROM. In this case, ROMs can be used to down-select candidate HEA compositions and to enable downstream characterization efforts.

B14. Develop reduced order models to study creep behavior
A specific area where ROMs could be particularly useful is in performing an initial assessment of creep effects in various multi-component systems. Initial ROMs could be based on existing creep deformation models, such as the Larson-Miller Parameter[196] and utilizing known information regarding diffusion.

B15. Develop and/or enhance oxidation models
This approach may involve the development of models to predict the process of oxidation (e.g., mass gain, thickness of the underlying alloy, and presence and thickness of oxide layers) from ab initio and/or thermo-kinetic modeling. These models could be used in conjunction with high-throughput experiments, for input to and guidance of full-scale experiments.

B16. Develop strengthening models
These would include both solid-solution and polycrystalline models for HEA systems, which also consider temperature effects. They should undergo verification and experimental validation (V&V), and uncertainty quantification (UQ) methodologies.[197,198] This preliminary recommendation can contribute to recommendation B12 above.

B17. Develop thermodynamic models for accurate prediction of phase stability and equilibrium
Understanding the phases present in potential new alloys is critical for the efficient design of next generation HEAs. CALPHAD methodologies supplemented with information from density functional theory calculations is a proven approach for this purpose.[199]

B18. Develop tools for calculating thermal, phonon, and electron conductivities
State-of-the-art tools in this area, see for example, need to be adapted to work for HEAs.[200,201]

B19. Code a new suite of ML algorithms designed for HEA discovery and design
Depending on the properties of interest, optimal machine learning (ML) algorithms and features (i.e., descriptors of the materials composition, microstructure, processing to be used as inputs) should be identified.

B20. Develop predictive tensile and high-temperature property computational models designed specifically for corresponding high-throughput HEA test methods
These simulations could include modelling strength, ductility, and oxidation resistance for HEAs slated for structural applications, e.g., refractory HEAs, light-weight HEAs, and/or ultrahigh-temperature ceramic HEAs.

B21. Develop and validate first-principles predictive models for HEA behavior in extreme conditions
This recommendation is integrated with a number of the other recommendations above. There is a need for developing computational tools that have been properly verified and validated with adequate uncertainty quantification for capturing the multi-component interactions and the nature of HEAs and properties. More specifically for this recommendation, such quantitative models are critically needed to supplement expensive and/or complex experiments performed in extreme conditions (e.g., high temperatures, cryogenic temperatures, high radiation levels, and/or highly corrosive environments).

C. High-Temperature Testing and Processing Equipment

C22. Establish rapid screening methods for HEAs in high-temperature environments (≥1300°C)
Screening methods should include examining the effects of temperature on elastic properties, strength, microhardness, thermal stability, and creep resistance.

C23. Develop standards for inductive and resistive ultrahigh-temperature (UHT) testing
It is important that the same standards be applied to all experiments so that correlations with existing UHT tests can be established.

D. Scattered Data with Uncertain Materials Pedigree

D24. Convene a broad HEA data consortium or working group
The aim of this body should be to: (a) define consensus-based HEA data ontology and schema, (b) identify or establish data storage platforms that address the needs of the HEA community, and (c) create an access-controlled Application Programming Interface (API) to permit access to all HEA data (e.g., analogous to The Materials Project (materialsproject.org)).

D25. Identify high priority HEA systems for which to generate substantive pedigreed materials information

The resulting information should be made available to participating research teams in accordance with the FAIR (Findable, Accessible, Interoperable, Reusable) guiding principles for scientific data management and stewardship.[202] It is imperative for both researchers and funders to implement and support the higher standard data infrastructure which pedigreed data enables.

E. Fundamental Composition-Processing-Microstructure-Property Knowledge

E26. Use artificial intelligence (AI) algorithms and other data informatics approaches to support composition-processing-microstructure-property relationships

Activities of interest include employing: (a) natural language processing (NLP) to survey existing literature, (b) machine learning algorithms to analyze composition and processing metadata, and/or (c) computer vision techniques to examine microstructural information obtained via microscopy and/or X-ray/neutron diffraction techniques. The precision offered through an adaptive, iterative AI approach, when used to inform high-throughput testing, allows the community to achieve targeted theoretical compositions, structures, and properties with limited initial screening.

E27. Expand knowledge of new HEA systems and processing conditions

Designate processing facilities as shared user facilities that focus on the various HEA processing methods needed to develop new HEA systems that use combinations of principle elements, or concentrations of those principle elements, that have not yet been studied. The resulting output should generate pedigreed composition and processing data that is housed in FAIR-principled[202] databases.

F. In Situ Characterization Methods

F28. Develop and designate national user facilities

Such facilities should target performing informed, high-throughput experimentation of HEAs that leverages in situ characterization measurements. These capabilities will allow for examination of the time dependent evolution of microstructure and properties, as a function of alloy composition and processing parameters. In addition, the integration of data science and AI approaches will be integral to achieving “smarter”, closed-loop high-throughput experimentation. As an example, such future national user facilities could be modelled after some of the current centers in other technical domains that are supported by NIST, DoE, and/or DoD.

G. Thermodynamic Databases for Complex Concentrated Alloy (CCA) Combinations

G29. Develop CALPHAD databases for HEA systems

This activity would require a sustainable funding stream for robust development and validation, and subsequent socialization, promotion, and encouragement within the greater community. These databases would preferably be open source but, alternatively, could be commercially developed.

G30. Develop a data schema framework around thermodynamic databases

This would provide a more robust representation of complex HEA thermodynamics, possibly related to cluster variation methods and/or other analytical and visualization techniques.

G31. Populate new HEA thermodynamic databases using experimentally established phase equilibria data for concentrated multi-component alloys
This effort would initially focus on experimental data down selected from promising HEA systems identified by predictive computational approaches. Expansion could then include more rigorous, high-throughput experimental data, as it becomes available for more HEA systems.

H. Availability of Affordable Feedstock Powder

H32. Establish a research-scale powder manufacturing facility for HEA development
This facility should be made accessible to multiple government agencies and associated research partners.

H33. Form a centralized feedstock facility or consortium group
This facility or group would function as a feedstock producer of small- to medium-batch ingots and powder lots. They would also pursue the development of non-traditional, low-cost, melt-less powder feedstock synthesis technologies.

I. Workforce Trained in HEA Exploration and/or Development

I34. Establish graduate-level internships and cooperative educational programs
These programs could offer a rotation of alternating opportunities across various HEA techniques, methods, expertise, and organizations.

I35. Create HEA-related tutorials
These tutorials could include a suite of hands-on, self-paced tutorials for HEA material data handling, ML-based analyses, and use of predictive computational tools. These materials should be easy to incorporate in existing classes and for self-study purposes.

I36. Produce a multi-institutional workshop series
The purpose of this series should be to provide students and early career professionals with opportunities to learn about specific HEA aspects and techniques from subject matter experts.

The HEA expert team was asked to provide a rough estimation of where these preliminary recommendations might fall on a chart of probability of success vs. impact. These results are provided in Figure 1 below but should only be viewed as additional, very subjective/qualitative input, rather than a strongly quantitative measure of projected success or impact. Nevertheless, readers might consider those preliminary recommendations in the upper right quadrant of Figure 1 to perhaps be ones that would represent potential areas of both high impact and high probability of success. Figure 1 was also taken into consideration during the development and prioritization of the detailed action plans and tasks in Section VII. It should be noted that the preliminary recommendations and Figure 1 represent only initial inputs that the team considered, along with the challenges and needs in Section V, as they deliberated further and fleshed out detailed, high-priority action plans in Section VII.

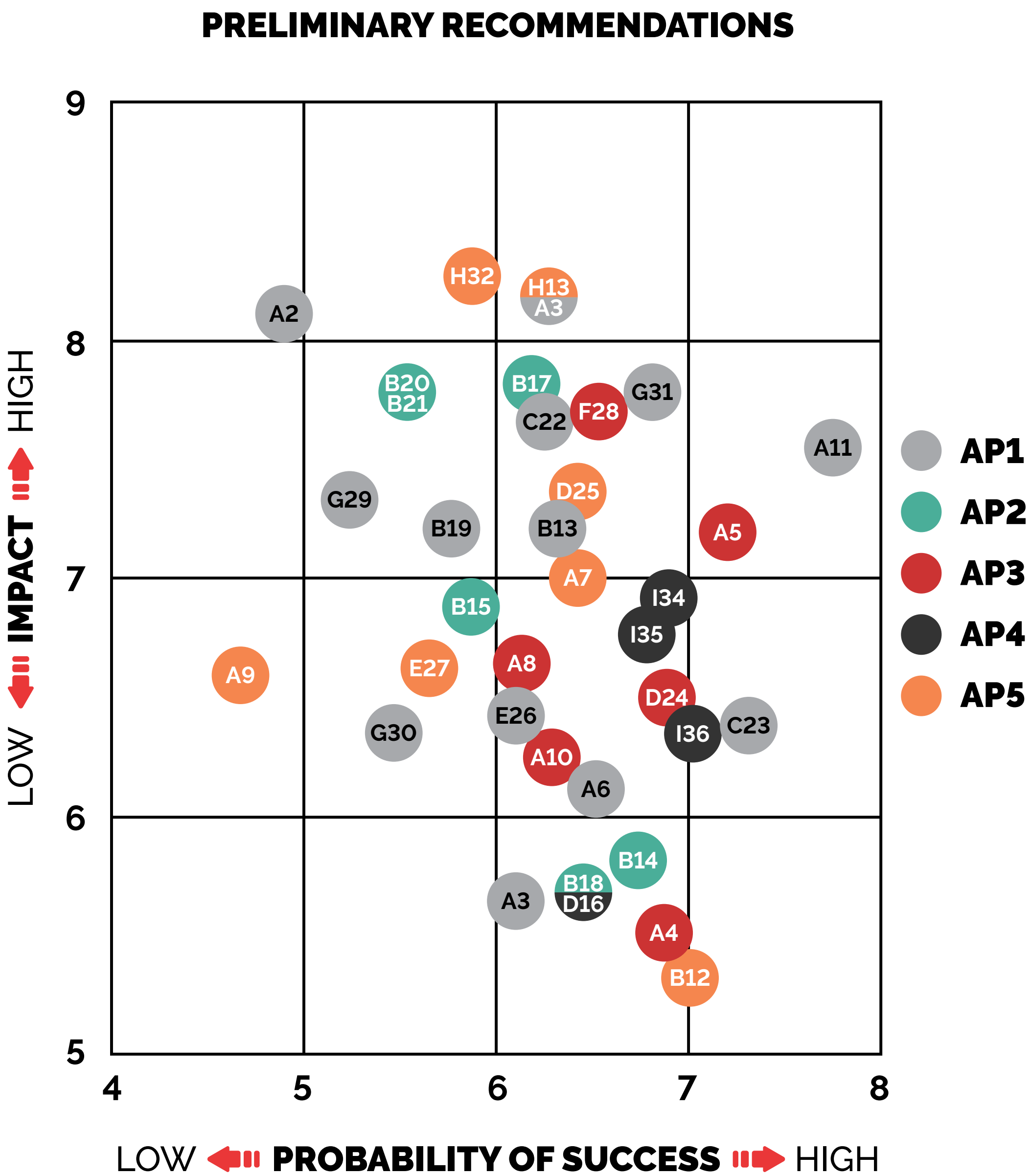

Figure 1: Preliminary recommendations plotted in terms of probability of success vs. impact (a subjective estimation). AP corresponds to “Action Plan”.

VII.

Recommended Priority Action Plans and Detailed Tasks

Using the challenges and needs (Section V) and preliminary recommendations (Section VI) as a foundation, the study team developed a set of five high-priority, detailed action plans to address some of the key needs and enablers concerning HEAs.

More specifically, the intent of these action plans is to provide more details to help scientists, engineers, and other stakeholders interested in HEAs accelerate the discovery, development, and implementation of these potentially game-changing materials. Detailed tasks/activities are provided for each action plan, and issues considered for each task include the recommended time frame for completion, milestones and/or progress metrics, estimated costs, and the types of key players and roles required. These action plans are synopsized below (in no specific order of priority), along with the related key tasks for each action plan (e.g., Tasks 1.1–1.8 for Action Plan #1).

Action Plan 1:

Develop High-Throughput Evaluation and Testing Methods

1.1 Develop a search strategy
1.2 Develop new synthesis methodologies for establishing HEA materials libraries
1.3 Develop post-synthesis thermomechanical processing techniques
1.4 Design an autonomous materials research (AMR) platform for HEAs
1.5 Establish high-temperature, high-throughput tests
1.6 Establish a database and common data schema and infrastructure for high-throughput HEA data
1.7 Establish active (extrapolative) machine learning methods
1.8 Integration and demonstration

Action Plan 2:
Develop or Extend Foundational Theory and Computational Models for HEAs

2.1 Develop or extend theory to accommodate multiple major elements
2.2 Extend theory and modeling of surface/subsurface thermodynamics and kinetics
2.3 Develop interatomic potentials for large-scale MD simulations
2.4 Develop ability to predict electron-phonon scattering lengths
2.5 Develop computational models for predicting behavior under extreme conditions
2.6 Generate input data for machine learning models
2.7 Develop validated ML models for phase and properties predictions
2.8 Develop phase-field modeling to simulate corrosion and oxidation

Action Plan 3:
Develop a Nationwide Network of Interconnected HEA Partnerships

3.1 Form a government agency working group, team, or oversight committee
3.2 Establish the incipient national network of HEA Centers of Excellence (CoEs)
3.3 Develop strong academic–industry–Government partnerships for HEAs
3.4 Launch a DoD-led consortium on high-throughput/autonomous research
3.5 Assign specific HEA topics to future CoEs
3.6 Launch an HEA CoE for theory, modeling, and data acquisition/management strategies
3.7 Launch an HEA CoE for technology transfer and scale-up
3.8 Launch an HEA CoE for design-quality test data collection, storage, analysis, and sharing

Action Plan 4:
Engage in Feedstock Production and Scalability Research

4.1 Establish a small-batch production facility
4.2 Identify existing powder production facility partners to aggregate feedstock demand
4.3 Introduce material provenance requirements for property databases
4.4 Provide researchers access to thermomechanical processing facilities
4.5 Research new powder production methods
4.6 Establish high-throughput screening protocols to access HEA processibility
4.7 Generate processing and property data for legacy and emerging HEA materials
4.8 Develop melt-less processes for ultrahigh-temperature materials
4.9 Conduct research on powder recycling issues

Action Plan 5:
Develop Training and Resources for Workforce Readiness

5.1 Organize courses and/or workshops on applicable computational techniques
5.2 Identify internship opportunities
5.3 Incorporate HEA topical foci into Materials Science & Engineering (MS&E) curricula
5.4 Develop HEA textbooks and/or chapters
5.5 Create certificates and/or credentials associated with short courses

Action Plan 1: Develop High-Throughput Evaluation and Testing Methods

This action plan is related to challenge area A from Section V—*High-Throughput Screening Methods and Experimental Tools*. It also integrates with the following preliminary recommendations from Section VI:

- Develop a set of high-throughput HEA test methods for measuring properties of refractory HEAs (RHEAs) and ultrahigh-temperature metallic and high entropy ceramics (HECs) (A1)
- Develop a closed-loop autonomous screening capability that integrates materials synthesis, characterization, and machine learning computational tools (A2)
- Establish data management and mining approaches (A11)

This action plan is motivated by specific needs and opportunities for the characterization of HEAs. When faced with the daunting size of the unexplored HEA composition space, researchers often struggle to identify where to begin. This conundrum establishes the need for a search strategy that quickly screens a vast number of candidates against selection criteria for a particular application and reduces the initial search space to a workable number of the most promising materials for further, more detailed examination. Such a search requires computational and experimental tools that can characterize essential properties quickly and unambiguously, under operationally relevant conditions, and to an acceptable level of precision. This also requires strategies that further accelerate exploration and screening by intelligently coupling computational and experimental characterization tools and the order in which they are applied. In general, some high-throughput methods are commercially available and reduced to practice for composition and microstructure, and computational and experimental screening methods have been validated for many key properties. Nevertheless, high-throughput characterization methods are still lacking for several key material properties, and most high-throughput characterizations have not yet been reduced to practice and integrated into a purposeful workflow.

In addition to barriers for high-throughput characterization of candidate materials, other challenges exist. Rapid and flexible material synthesis that can produce materials libraries consisting of either compositional or microstructural gradients (or both) are needed, along with an ability to produce arrays of discrete, "on-demand" alloys. Further, structural applications require bulk-like materials libraries, not thin films.[101] Additive manufacturing (AM) has been demonstrated for a select set of materials libraries, but additional work is required to produce the full range of necessary materials libraries in both metallic and ceramic materials. High-throughput methods are needed to rapidly evaluate the vast range of post-synthesis, thermo-mechanical methodologies used to control HEA microstructures (and hence properties). The lack of an established database structure and ontology makes existing data dispersed and non-interoperable. Given the different data modalities and data volumes, automated, web-based approaches for capturing, storing, and sharing data are critical. A more coordinated and automated approach would enable artificial intelligence (AI) tools, as well as machine learning and natural language processing algorithms, to accelerate data analysis and recommendations for subsequent trials.

Autonomous materials research (AMR) is a quickly growing field that integrates high-throughput evaluation and testing methods and the other steps described above into a closed-loop, unsupervised process to vastly accelerate materials exploration and innovation.[106] The AMR process as envisioned in this study is illustrated in Figure 2 on page 42, and described in the AMR callout box beginning on page 42. While significant strides have been made in AMR methods,[101] there is still much work to be done. AMR efforts typically optimize against a single objective function, while most applications require optimization against many criteria that often compete against one another. Autonomous research efforts are currently limited to materials that can be produced by vapor or solution chemistry, or by extruding polymers (see Table 1 in Stach et al.[106]). Structural materials offer dual, orthogonal complexities of composition and microstructure, adding a new layer to the AMR cycle. Inorganic structural materials offer daunting challenges in the synthesis and post-processing control of microstructure that are not currently addressed in efforts to advance the AMR method.

This action plan addresses two major needs found in this study—the development of HEAs for high-temperature applications and the acceleration of HEA exploration—by overcoming present gaps in the AMR concept and applying them to Refractory HEAs (RHEAs) and High Entropy Ceramics (HECs). The successful completion of Action Plan 1 is expected to produce and validate an AMR capability for high-temperature structural applications. This provides the foundation needed to apply AMR methods to a much broader range of materials and applications than is currently possible, advancing not only the HEA field, but the broader field of materials science as well.

Task 1.1–Develop a search strategy

This task considers the fundamental steps in the autonomous exploration of a vast search space of inorganic, high-temperature structural materials, and establishes the principles to organize these steps for maximum speed and minimal use of resources (time, materials, facilities, cost). This task begins with a study to evaluate key considerations and decision points within the autonomous materials research (AMR) process. These considerations are posed as questions and discussed briefly below. *For a given application, what are the minimum essential characterizations needed to support a down-select decision during the screening process?* While the specific characterizations will be unique for each particular application, there are likely a set of common principles that can be used to evaluate each unique case. The purpose of this study is to determine these principles. *What tools are best used for each characterization?* A material can be characterized by computations or by physical measurements. Computations include physics-based models, numerical simulations, and phenomenological or empirical methods. Important considerations in deciding which tool to use include the resources needed and the accuracy or uncertainty provided. *In what order are the characterizations best performed?* The order in which the necessary evaluations are done matters a great deal—those that can eliminate the largest number of candidates with fewest resources are best done first.[194] Further, some tests are destructive; these must be conducted last. Methods to define the resources and uncertainties associated with different characterization methods are needed. *How are test conditions and pass/fail criteria defined?* Rapid screening tests may often be performed under conditions that do not exactly match the operational environment of an application, and so careful thought is needed to define both the parameters used in the characterization (for example, exposure time, temperature, and gaseous species for oxidation resistance) and the pass/fail criteria. Again, the goal of this task is to establish the underlying principles needed to determine these parameters and criteria for a wide range of applications rather than for a specific application.

Risk of uncertainty also needs to be considered in this topic. *What information is collected?* This information is essential so that AI and ML tools may extract the maximum information from subsequent analysis of the characterizations performed. The type and amount of data collected also has important implications for storage, retrieval, and provenance. Careful consideration of the guidelines used to establish the minimum data needed to support these functions will be performed in this task. *What criteria are needed to support an unsupervised decision to continue or to exit the search loop?* This decision is an essential component of autonomy in the AMR process. At least three different outcomes are possible at this decision point: continue the autonomous search by initiating synthesis and evaluation of the next campaign suggested by the AI/ML modules; consult with the human-in-the-loop; or exit the search loop, thus completing the search.

Answers to the above questions may be pursued by conducting a study of previous work on relevant topics. This includes efficiency studies in engineering, industry, and management, and will consider methods such as sampling techniques, iterative design, and design of experiments to extract the maximum amount of actionable information from the minimum number of characterizations. Decades of experience in the combinatorial exploration of functional materials may also provide useful guidance. Industrial knowledge is also essential, especially regarding the process of decision-making associated with design criteria and quantifying the risk associated with decisions. Finally, this task will develop deeper insights by applying the knowledge gained from the studies above to design a specific search strategy to explore Refractory HEAs (RHEAs) and high entropy ceramics (HECs) for two-to-three DoD-relevant high-temperature structural applications. The estimated time and cost to accomplish this task is 1 year and $1M (Note: this and all subsequent monetary values are provided in USD).

Task 1.2–Develop new synthesis methodologies for establishing HEA materials libraries

This task focuses on building infrastructure to synthesize HEA samples in real time as they are identified through the unsupervised ML recommendations from the AMR process. Rapid, automated synthesis for establishing HEA materials libraries introduces new challenges to processes such as additive manufacturing (AM). A large number of AM powder feeds may be required, along with the fundamental understanding needed to control the transport rate for powders of different densities, particle sizes, and powder shapes. Assuming the existence of a high-throughput database (see Task 1.6), the first milestone for this activity may be to identify and document process-specific challenges that are unique to high-throughput synthesis. A particular challenge for AM is the selection of optimum process conditions, including powder flow rates, laser power, scan strategy, and scan rate for any arbitrary experimental alloy. Initial research efforts are already underway on this topic;[203] this task builds on that foundation and represents a more intense, focused effort. Additional challenges addressed by this task are development of process conditions needed for AM of RHEAs and HECs with very high melting temperatures and sensitivity to contamination from interstitial elements. Practitioners should document challenges and solutions when modifying process conditions for different alloying elements within a single HEA materials library, taking advantage of known Integrated Computational Materials Engineering (ICME) methodologies and tools[89–91] to further expedite the synthesis of specific materials.

Benefits will arise from a collaboration of academic, industrial, and national laboratory participants to develop new synthesis techniques and platforms, as well as partnering with industry to obtain funding opportunities (possibly through Small Business Innovation Research (SBIR) grants) to develop necessary equipment. Moreover, primary metal processing experts may be consulted to guide synthesis development and help ensure that materials libraries are representative of larger specimens, parts, and/or components of interest.

These task objectives may be met with three years of dedicated, collaborative work supported by funding on the order of $5M. Successful completion of this task may be considered to be achieving a final goal of producing more than 10 HEA sample alloys per day (per high throughput synthesis unit), with compositional and microstructural control representative of material produced by more conventional, larger scale methods.

Task 1.3–Develop post-synthesis thermomechanical processing techniques

To ensure that measured properties are not limited by testing inappropriate microstructures, post-synthesis techniques such as heat treatments and deformation processing (together, called thermo-mechanical processing, TMP) may be needed for many materials libraries. This is an untouched domain—there are no studies that explore the TMP of a host of different compositions embedded within a single HEA materials library, to control microstructure. This task requires significant original work to develop new concepts, new scientific foundations, and new practical approaches capable of producing controlled microstructures in a library that spans a range of distinct alloy compositions. Specialized facilities may be required to enable high-throughput TMP of multiple specimens in parallel, and so involvement of national laboratories with experimental user facilities may be appropriate. These facilities may include novel dies and fixtures with adequate properties to enable TMP of HEAs at temperatures and stresses appropriate for high-temperature structural materials. Teaming is expected to include academia to establish the necessary scientific foundations, industrial partners to produce custom dies and fixtures, and national laboratories as mentioned above. Innovative concepts essential to this topic may come from any contributor within the entire materials enterprise. This activity is expected to require a minimum of 2 years and an estimated cost of roughly $3M.

Task 1.4–Design an autonomous materials research (AMR) platform for HEAs

The objective of this task is to design a robust platform for the high-throughput measurement of HEA properties. Most high-throughput experiments for structural materials have been validated, but these tests are typically demonstrated with a single sample size, shape, and/or geometry (i.e., sample "form factor"). To advance this technology, modifications are needed to each of these tests to accept a materials library of either continuous gradients or an array of discrete samples. Design alterations are also needed to maximize automation and robotics that can quickly perform repetitive tasks and transfer the materials library between stages. This requires the design of a standardized kinematic sample holder that can move from synthesis and post-synthesis TMP to each of the physical characterization stages. Such a holder should be compatible with a suite of optical and electron microscopes and multiple physical testing devices (see Task 1.5 for more details). Further, the experimental methods must be designed to run evaluations on every sample in a library simultaneously, or to employ automation and robotics for sample translation when tests on a library are performed in series.

To scope and constrain the design space for an adaptable AMR system, investigations may commence by considering high-throughput characterization methods that are already validated. As a major contribution, this task will also develop essential, missing characterization methods that are likely to have high impact across a range of materials and application classes—for structural materials this includes for instance melting temperature, tensile ductility, and environmental resistance. A major objective of this action plan is also to extend existing high-throughput tests to higher temperatures, this is discussed separately in Task 1.5. Additional constraints from these newly developed tests will be included in the design of a robust, adaptable AMR system. This design effort will consider the cost of facilities, test sample/materials library requirements, time per evaluation, capability for elevated temperature testing (see Task 1.5) and the quality of data produced.

To apply the design concepts developed in this task and to demonstrate reduction to practice, this effort will design and construct an integrated AMR system to explore RHEAs and HECs for the high-temperature structural applications identified in Task 1.1. Given the breadth and difficulty of this task, this study estimates a budget of ~$10M over 5 years to adequately address each of the activities. In addition to widespread contributions from academia and government laboratories, Federal agencies such as the National Institute for Standards and Technology (NIST), agencies with the Department of Defense (DoD), and the Department of Energy (DoE) may be logical partners to benchmark specific properties of interest. Industrial collaboration is considered essential for this task to include extensive experience in the design of effective materials search systems and to ensure the proposed AMR design concepts are compatible for operation within an industrial setting. Industrial experience in the areas of automation, controls, and robotics may be especially valuable. The ability to produce and market specialty equipment associated with an AMR workflow may further support not only industrial collaboration, but also the formation of new, small businesses.

Task 1.5–Establish high-temperature, high-throughput tests

Development of HEAs for use in harsh environments (e.g., high temperature, corrosive, nuclear, cryogenic temperatures) is an area of great promise, and active research. To make this task tractable, only HEAs for high-temperature environments will be considered in this task. In this regard, the HEA field, through RHEAs and HECs, offers a particularly strong motivation to develop new high-temperature structural materials. High-throughput experiments that can be conducted at operationally relevant temperatures are required to evaluate RHEAs and HECs for this broad class of applications. Current Ni-based superalloys extend to nearly 1100°C, and dramatic benefits are certain for materials that can support engineering loads at temperatures of 1300°C and above. Commercially available testing equipment that can operate at these temperatures is limited, and essentially no equipment is currently available to perform such tests in a high-throughput mode. This equipment must produce and measure the desired temperatures, apply appropriate loads or forcing functions, and measure the material response in a vacuum or inert environment to minimize sample damage. Specialty heating methods, environmental chambers, loading devices, measurement sensors and devices, and controls for thermal stability that can withstand the temperatures of interest are all required. Developing these specialty components into working systems and making them available will require significant industrial and academic collaboration.

A few selected tests are mentioned here to help focus considerations on this task. The most important properties for high-temperature structural materials are melting temperature (synthesis, TMP, and a wide range of properties scale with melting temperature), room temperature tensile ductility, strength at the maximum use temperature, and environmental resistance.[101] Indentation methods can provide an acceptable evaluation of strength,[204] but the ability to conduct such tests at elevated temperatures, especially as high as 1300°C, remains elusive. Innovative tensile testing approaches that automate sample preparation, transport and loading have been reported and show promise but still require improvements in handleability, reduced data acquisition time, and extension to elevated temperatures. The small punch test (SPT) has also been validated but will benefit from improved reliability and engineering modifications to enable the automated testing of many samples from a single materials library and, once again, use at high temperatures. Nearly half a dozen different approaches have been reported to study high-throughput oxidation, but each has limitations that need to be overcome to provide a more general method for high-temperature structural materials. It is easy to locally melt a sample with a laser, but the ability to measure the temperature of an arbitrary alloy without prior calibration to account for conductivity and reflectivity remains the primary barrier to a rapid, general method to measure melting temperature. Other high-throughput tests that may be important for high-temperature structural materials include creep and fatigue. As an important constraint, strength, tensile ductility, and environmental resistance of high-temperature structural materials must be measured on bulk-like samples—properties measured on thin films have very limited applicability. All the high-throughput test methods developed in this task should pursue the use of automation and parallel sample machining, handling, and testing to the maximum extent possible. This task provides a significant opportunity for creativity and innovation.

Given the need to develop novel equipment suites and testing methodologies, it will likely take on the order of 2–3 years and $2–3M dollars to develop working systems for each of the following high-throughput tests: (1) high-temperature hardness, (2) tensile ductility, (3) oxidation screening, and (4) melting temperature. Therefore, this activity might require a total of ~$6–10M dollars over anywhere from 2–10 years, depending on how many of these projects are undertaken in parallel. All parts of the science and engineering enterprise may provide important contributions to this task, and strong involvement from small and large businesses may help reduce these methods to practice and make systems commercially available for more widespread use.

Task 1.6–Establish a database and common data schema and infrastructure for high-throughput HEA data

As the amount of HEA high-throughput modeling and testing data increases, it is natural that various databases and data schema will emerge to house and organize this information. However, such structures can quickly become unwieldy or obsolete if not thoughtfully constructed. Construction of a successful database requires long-term investment, broad participation, and community-wide acceptance of data capture methods and storage methods that enable seamless data sharing. Therefore, it is vital for community stakeholders to develop a database schema that both captures and relates pertinent metadata regarding (1) composition, (2) synthesis and thermo-mechanical processing techniques, (3) microstructural features, (4) structural and functional properties, (5) high-throughput characterization method used, (6) elemental material costs, and (7) recyclability and environmental behavior, amongst others. Due to the complex and variable nature of HEA systems and potential processing techniques, it is imperative that the ontology remain as broad as possible to accommodate input from a wide range of HEA systems and/or relatively uncommon HEA processing methods (e.g., ball milling). This database should also have the capacity to document data from undesirable, non-stable systems that were explored, so it can be used to better train AI-assisted search strategies.

An initial milestone in accomplishing this task is to outline and execute a quantitative time-based strategy to input existing legacy data (for a predetermined number of alloys) into the database. This activity would take approximately $1M in seed funding as well as ~ 9 months of coordinated effort between federal agencies, professional societies, and the community at large to develop a data schema that can effectively capture and organize legacy data while also being sufficiently flexible to grow with the needs of this still-evolving field.

Task 1.7–Establish active (extrapolative) machine learning methods

To expedite the systematic search of vast, non-dilute computational space, it is critical to leverage the power of artificial intelligence (AI) techniques such as machine learning (ML) to search for impactful HEA systems. Machine learning algorithms offer great potential to guide material discovery through extrapolative means. By employing simulated or measured experimental datasets to predict potential regions of interest, ML methods can greatly reduce the amount of subsequent computing time and physical experimentation needed to find potentially interesting HEA systems for further, detailed exploration. Once trained, these algorithms can be refined for either single-objective or multi-objective optimization. A first milestone for this activity is to develop an ML algorithm with batch-sized learning methods which implement established techniques for single-objective optimization. As this algorithm is refined, it can be augmented to accommodate new multi-objective optimization approaches as well. The study team envisions an effort of ~2 years and $1M for academic and national laboratory stakeholders to develop such an algorithm. In addition, industrial and governmental entities would need to be engaged to provide access to databases for model training and benchmarking.

To take full advantage of these ML techniques, a second milestone is to embed physics-based models to improve extrapolative modeling results. This would require considerable effort on the part of academic and national laboratory stakeholders to develop fundamental theories that are applicable to non-dilute materials systems (see Section V Subsection B for more discussion).

Task 1.8–Integration and demonstration

The final task in this action plan is to integrate the advancements made in each of the preceding tasks into an operational AMR capability, and to validate this system by applying it to 2–3 selected materials design challenges. As mentioned earlier in this action plan, to start with a tangible goal, the selected materials challenges recommended in this section will first apply RHEAs and/or HECs to high-temperature structural applications of high DoD impact. But some other also critically important AMR capabilities to be developed could include those centered around corrosion, nuclear/radiation, cryogenic, and/or biological application areas as well.

As part of this task, the reliability and stability of the final search results may be evaluated against the influence of uncertainties, the chosen pass/fail criteria, and false positive/negative results. In addition, best practices and suggestions for improving future search strategies can be performed and documented in a manner congruent with previously published verification and validation methodologies.[197,198] It is estimated that, given the accomplishments (and related investments) from Tasks 1–7, an initial instance of a search strategy for 2–3 key applications as described in this final task could be developed for approximately $1M over the course of a year. This activity would require input and contributions from key industrial, academic, and governmental stakeholders.

High entropy alloys (HEAs) bring challenges to the autonomous materials research (AMR) field that do not seem to be addressed by current AMR efforts. Here, we broadly interpret the AMR process and modify it to include these new challenges. A flow diagram of the AMR process for HEAs is shown in Figure 2. Each of the stages are then briefly described below this figure.

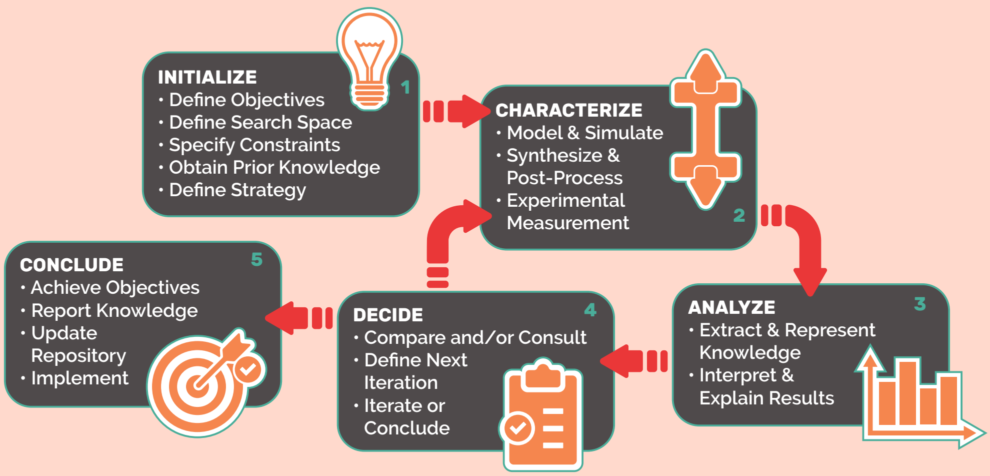

Figure 2. Illustration of the autonomous materials research (AMR) process. This cycle includes artificial intelligence (AI) and unsupervised machine learning (ML) algorithms to: characterize materials via simulations and physical experiments; analyze results and propse new materials for subsequent evaluations; and initiate and conduct subsequent evaluations without human intervention. A decision point is also included that can trigger consultation with the human-in-the-loop, giving a synergistic interaction between human creativity and insights, and AI/ML contributions. This consultation also gives an opportunity to interact with the component design function, providing more acenues for innovation. This AMR process is based on the model proposed in Stach et al.,[106] and is adapted for HEAs and other bulk, inorganic materials using concepts from Miracle et al..[101]

In ***Initialize***, the objectives of the AMR process are defined by selecting an application and consulting with the design function to identify the minimum set of properties needed to select a promising or fully successful candidate (see *Conclude* below for a discussion of the difference between these two sub-tasks). The palette of elements included in the search space may also be defined, along with the microstructures to be considered. Prior knowledge is established by collecting available relevant data and organizing the data into a usable format.

The strategy to be used in the AMR process is an essential feature of the *Initialize* step. The strategy includes definition of the high-throughput methods that will be used to characterize each of the properties evaluated, including both modeling and simulation (M&S) tools and physical measurements (experiments). The characterization methods may not be able to reproduce the exact service environment, and so the conditions used in the characterizations (both M&S and experimental) are defined, along with the pass/fail criteria for each test. The conditions for success of the AMR process are also defined in the strategy. An AMR search will typically consider multiple objective functions, and some of these may display inverse or competing trends with parameters such as composition or temperature. Thus, the definition of success may include a minimum value for each objective function, may define a minimum position achieved by the Pareto front[a] of the combined objective functions, or may consider some other method to optimize material response across the different objective functions. Finally, the strategy defines the candidate alloys to be evaluated in the first iteration of the AMR process. The *Initialize* stage is done with a dominant contribution from the human-in-the-loop.

In ***Characterize***, the initial candidates identified in the search strategy are evaluated. Here, the term 'characterize' includes the use of both M&S (computational) tools and physical measurements (called experiments). A single iteration in the *Characterize* step may include two or more layers of evaluations. M&S is first to be performed, since computations will generally eliminate the largest number of candidates with the least amount of resources (time, material, facilities). These tools can include atomistic calculations, physical or numerical models, and phenomenological correlations. Because M&S tools are not available for all desired characterizations, experiments are also needed. Only those candidates (compositions and/or microstructures) that pass the M&S characterizations are synthesized and tested in the following high-throughput experiments. Materials libraries for candidates that pass the M&S stage are produced autonomously. If specified in the *Initialize* stage, post-synthesis, thermo-mechanical processing may be applied to the materials libraries to control the microstructure or reduce defects. High-throughput experiments are then performed on the materials libraries. Properties that depend primarily on composition are evaluated first and may use materials libraries where composition is the only variable. Properties that depend strongly on both composition and microstructure (such as strength and ductility) may require additional layers of characterization (and additional materials libraries) to evaluate variations in both composition and microstructure. Alloys that fail to pass initial screening experiments need not be characterized in subsequent layers of evaluations to save time.

a. In the space of objective functions in multi-objective optimization problems, "Pareto front" generally refers to a set of optimal solutions that are non-dominated to each other but are superior to other solutions in the search space. In other words, it is not possible to find a single solution that is superior to all other solutions with respect to all objectives, such that each solution of the Pareto set includes at least one objective inferior to another solution in that Pareto set, although both are superior to others in the rest of the search space.[205–207]

The ***Analyze*** step in the AMR process extracts data and metadata from the characterizations. The metadata includes not only the compositions and microstructures tested, but also the synthesis method and post-synthesis thermo-mechanical processing conditions, the characterization method used, and the parameters needed to reproduce this characterization. The results are interpreted by applying physical and phenomenological models to the extracted data, including ML algorithms. The results are further analyzed to compare against the definition of success for the search. Finally, recommendations of candidate materials for subsequent iterations are made using unsupervised ML and AI methods.

In ***Decide***, the AMR process autonomously compares the characteristics of the current set of candidates against the criteria for success defined in the *Initialize* stage. If all criteria are met, the process moves to the *Conclude* step. If one or more criteria are not satisfied, then an option to consult with the human-in-the-loop can be made. This consultation can include both the materials and design functions, with the intent to allow adjustments to the AMR cycle. Based on accumulated knowledge from previous iterations along with human creativity and insights, the materials function may: (1) adjust the scope of the search by changing the palette of elements considered; (2) change the test conditions applied in the characterizations; or (3) adjust the pass/fail criteria. The design space often represents a level of complexity equivalent to the complexity of the materials. Consulting with the design function therefore offers human creativity and insights on adjusting the design requirements to reduce the search risk or accelerate achievement of the desired outcome. These consultations with the human-in-the-loop from the materials and design domains enable a synergistic interaction between man and machine. The *Decide* step ends with the decision to continue to the next iteration or to conclude the search.

The final step is ***Conclude***. The knowledge gained from the AMR process is reported and data repositories are updated. The AMR process is expected to provide actionable data, so that implementation is an essential follow-on activity. In some cases, the AMR result may be sufficient to support transition of the selected material to the intended application. In other cases, additional evaluations may be necessary, such as conducting extensive, standardized characterizations to certify a new alloy and to support specification of statistical design allowables. For example, initial screening for a high-temperature turbine blade material may evaluate room temperature tensile ductility, elevated temperature strength, density, stiffness, oxidation resistance, and cost. Successfully meeting these criteria may reduce an initial search space from many millions of compositions to a few dozen candidates. In this case, implementation may include further evaluations of the small number of successful candidates produced by the AMR process. This subsequent work may include the use of higher precision, standardized tests needed for alloy certification, for which high-throughput evaluations either have insufficient accuracy or statistics or are not presently available.

Action Plan 2: Develop or Extend Foundational Theory and Computational Models for HEAs

This action plan is primarily related to challenge area B from Section V—*Predictive Models and Computational Tools*, but also contributes to challenge area C—*High-Temperature Equipment and Testing*. It also dovetails best into the following preliminary recommendations from Section VI:

- Develop and/or enhance oxidation models (preliminary recommendation (B15))
- Develop tools for calculating thermal, phonon, and electron conductivities (B18)
- Develop predictive tensile and high-temperature property computational models designed specifically for corresponding high-throughput HEA test methods (B20)
- Develop and validate (with uncertainty quantification, UQ) first-principles predictive models for HEA behavior in extreme conditions (B21)

Prior to considering the detailed tasks for this action plan, it is useful to elaborate on the challenges and opportunities that provide motivation and justification for this specific action plan. By way of opportunity, the foundational theory and computational modeling considered here are critical for the accelerated development and implementation of HEAs. However, it is challenging to model the large number of possible local compositions and the myriad of possible microstructures and defects (i.e., with respect to chemical complexity coupled to structural complexity) in HEAs. Several challenges associated with linking various time and length scales will also need to be addressed for achieving process and engineering-scale HEA models.

Additionally, a key requisite for accurate predictive computational models is the availability of large, reliable experimental datasets for proper development and validation of such models. Along these lines, there is also currently insufficient HEA data for using brute-force machine learning (ML) approaches. One specific example is in applying active learning ML algorithms to predict phase stabilities. There is also a lack of validated interatomic potentials for large-scale molecular dynamics (MD) simulations related to HEAs, and current thermodynamic and kinetic databases for HEAs are not sufficiently reliable as well. Additionally, most commercial databases are encrypted/proprietary.

Moreover, oxidation and corrosion involve many stages with numerous elementary steps, yet selected stages have gaps in theory, lack of fundamental data, and lack of understanding regarding how multi-component systems operate with 3–6 major elements. Understanding is currently limited to 1–2 major elements. Additionally, local fluctuations in properties (due to chemical complexity) at the nanoscale (and/or higher scales) are not fully characterized, yet they affect response (e.g., dislocation mobility, oxidation, vacancy concentrations). In the realm of mechanical properties, there is a lack of reliable theory and models to predict temperature-dependent yield strength and ductility in HEAs.

Task 2.1–Develop or extend theory to accommodate multiple major elements

This activity would extend theory from 1–2 major elements to 3 or more major elements and include the concomitant structural and chemical complexities. It would build a foundation to support the ability to predict and master the engineering of "cocktail effects",[24] given the complexities involved. It is emphasized that this task refers to enhancing the underlying fundamental theories, equations, and mathematics that support computational models, rather than the development of the specific computational models or simulations themselves. Topical (interrelated) theoretical domains associated with phase formation could include thermodynamics, driving forces, phase stabilities, kinetics, transformation mechanisms, and triggers. In this context, triggers refer to nucleation and/or growth promoters such as pertain to corrosion, passivity, grain boundaries, and/or dislocations. Foundational properties-based theoretical treatments for HEAs would be equally important, such as those related to mechanical properties (e.g., strength, ductility, fracture toughness, fatigue, creep), extreme conditions (corrosion, high-temperature oxidation, radiation), and/or functional properties (e.g., magnetic, electronic).

The timeframe for this task could span a range from 2–10 years, depending on whether the theory is extended from existing materials science knowledge and theories or if larger gaps exist. A metric of success could be whether the extended/new theory is adopted by modelers and used as a foundational underpinning for predictive computational models and simulations that are properly verified and validated by experiments. Another metric is whether those computational models are then used to extend significantly the existing HEA database, and/or provide new ability to predict and master the engineering of complex "cocktail effects" or property extremes in HEAs. The roughly estimated cost to extend or develop new theories in this regard is perhaps on the order of 3–4 principal investigator (PI) grants for 5 years, at $150,000 per grant per year (~$3M total), for a given property or theoretical domain. The costs could also be dependent on the type of property or theoretical domain addressed. For example, some properties might be amenable to understanding via rules of mixtures, and thus would entail less costs for extending or developing the underlying theoretical basis. Others may depend on extremes, defects, and/or cocktail effects requiring wholly new theoretical treatments, such that developing the proper theoretical underpinnings will be more expensive. The types of personnel required for this task would include scientists at universities and/or national laboratories with expertise in theory (and any requisite mathematics), within application domains including physics, materials science, thermodynamics, and/or mechanical properties. Computational modelers and experimentalists would also be required to employ the theory in computational simulations, conduct verification and validation to help test the efficacy of such theories, and supply data where key gaps exist.

Task 2.2–Extend theory and modeling of surface/subsurface thermodynamics and kinetics

This task would primarily be concerned with extending the underlying theory and/or computational modeling of the thermodynamics and kinetics of surfaces, interfaces, and altered layer materials to the chemically and structurally complex systems associated with HEAs. It would encompass the development of working theories and models of fundamental unit processes and states, to be used for unanticipated combinations of elements. This activity would also likely involve *ab initio* methodologies. Experiments are required in parallel with computations for proper verification, validation, and uncertainty quantification of the associated computational models.

Costs are roughly estimated at a minimum of 2–3 PI grants, at ~$200,000 per year for each PI/grant. Key players would predominantly be researchers and engineers at universities and/or national laboratories. Proper development of these theories and computational models could take anywhere between 2 and 5 years, depending on the levels of complexity, functionality, and funding support.

Task 2.3–Develop interatomic potentials for large-scale MD simulations

This task addresses a need for increased accuracy of interatomic potentials that are transferable between bonding environments for application to HEAs. These potentials should be experimentally validated to quantify their accuracy. Some metrics of success related to these validations could include the demonstrated ability to predict melting temperatures and/or mechanical properties (such as single crystal strength). Coupling with Monte Carlo/molecular dynamics (MC/MD) techniques to simulate oxidation behavior is another example of a possible success metric. Associated costs for this activity would be on the order of $200,000 annually, say for two research groups/PIs, over 3–5 years (i.e., approximately $1–2M total). The types of researchers required for this task would include some combination of experts in electronic structure calculations associated with MD simulations. Additionally, people with expertise in machine learning and/or and optimization could contribute to this activity.

Task 2.4–Develop ability to predict electron-phonon scattering lengths

This task involves investigation of electron-phonon scattering lengths within highly disordered lattices, including defects such as vacancies. Examples of metrics of success would be to match experimental electrical and/or heat transport measurements across compositions and material types. Associated costs for this activity would be on the order of $200,000 annually for one or two research groups/PIs, over 3–5 years (i.e., anywhere from ~$0.6–2M total). This activity can be accomplished by researchers at universities and/or national laboratories with skills and experience in the atomistic approaches required for these types of calculations. Scientists and/or engineers with the ability to acquire the appropriate experimental data for the validation of the calculated results will also be required.

Task 2.5–Develop computational models for predicting behavior under extreme conditions

The types of models developed under this task would lead to simulation of ultrahigh-temperature oxidation, melting temperature, corrosion, and/or radiation behavior. These simulations can be used to supplement expensive and/or complex experiments and must incorporate uncertainty quantification methodologies. Examples of potential measures of success of this task are demonstration of the ability to predict melting temperatures of refractory alloys, and/or the ability to predict radiation damage. Quantitative measures of output and success could include number of publications and related citations. Costs associated with this activity would also be on the order of grants of $200,000 annually, for one or two research groups/PIs, over 3–5 years (i.e., anywhere from ~$0.6–2M). This type of activity would be accomplished by computational researchers from academia, government laboratories, and/or industry, working in conjunction with experimentalists for validation of the models.

Task 2.6–Generate input data for machine learning models

This activity would include developing and/or employing some combination of the appropriate theoretical foundations, computational models, and low-fidelity experiments needed (e.g., see Tasks 2.1–2.5 above) to acquire necessary input data for machine learning (ML) models. The overall objective of this task is to address the problem of the relatively scarce amount of HEA data within any given alloy or alloy family. There is a need for such robust datasets to build viable, valuable, ML models. Uncertainty quantification of the data and models within this activity is essential. The ML models could be used to develop descriptors (both theoretical and experimental) related to mechanical properties, functional properties, chemistry, etc. For example, "entropy forming ability" is a descriptor derived from first principles calculations of the energies of high entropy carbides that can predict the ease by which compositions can be synthesized.[208]

A measure of success of this task could be demonstration that the ML models which use this data are appropriate for active learning (i.e., iterative supervised learning) activities that support discovery, development, and/or implementation of HEAs. It is estimated that two or three research projects/PIs would be needed to support this project, at a level of funding of about $300,000 per property addressed. Scientists and engineers in universities, national laboratories, and/or industry would undertake this activity, and it would most likely require some personnel with expertise in applied mathematics.

Task 2.7–Develop validated ML models for phase and properties predictions

This task is directly related to Task 2.6. More specifically, it is important to develop theoretical ML descriptors for predicting phase stabilities that go beyond thermodynamic approaches. Here, ML models could be used to predict cocktail affects for fundamental properties (such as band gaps). Experimental validation of properties across materials classes and compositions would be a measure of good success of this activity. Accomplishment of this task could take anywhere from 3 to 10 years, depending on the number of materials classes and compositions for which these ML models provide validated predictive capabilities. To cover a significant range of compositional families might require consortia of computational and experimental groups, working collaboratively. This would best be framed by the development of HEA Centers of Excellence (see Action Plan 3 below) that encompass multi-institutional efforts, including collaborators from academia, government laboratories, and industry. To accomplish the full range of this activity could require large levels of support, perhaps on the order of $10M.

Task 2.8–Develop phase-field modeling to simulate corrosion and oxidation

These phase field models would simulate oxidation and corrosion of complex HEA materials, incorporating surfaces, surface oxides, and bulk material. This work would be conducted in parallel with density functional theory (DFT) calculations that are validated by experiment. A measure of success would be experimental validation of properties across multiple materials classes and compositions. Some quantitative metrics of success could include a number of publications, related citations, and/or invited presentations. The cost of this activity would be on the order of $200,000 annually for each of two research groups/PIs, over 3–5 years (i.e., anywhere from about $1–2M total).

Action Plan 3: Develop a Nationwide Network of Interconnected HEA Partnerships

This action plan is especially related to challenge areas A—*High-Throughput Screening Methods and Experimental Tools*, D—*Scattered Data with Uncertain Materials Pedigree*, and F—*In Situ Characterization Methods*, from Section V. In this vein, it also dovetails particularly well with the following preliminary recommendations (from Section VI):

- Increase coordination among government agencies (A4)
- Establish a DoD-led consortium group (A5)
- Create a mechanism for using shared high-throughput testing resources (A8)
- Develop a facility to conduct parallel high-throughput tests for strength, toughness, and creep (A10)
- Convene a broad HEA data consortium or working group (D24)
- Designate national user facilities (F28)

Despite the novel nature, growing interest, and revolutionary potential of HEAs, there are currently no nationally coordinated, multi-agency supported efforts among key academic, industry, and government groups on HEA development within the United States. The vast majority of HEA-related R&D has been limited to the work of individual research groups, with a few exceptions, including some multi-team DoD and NSF programs. This limited teaming has resulted in slow, sporadic progress, and a tendency toward deep investigation of known HEAs (such as the Cantor alloy), rather than coordinated searches for new and impactful HEA systems. This lack of coordinated investment has led to the US lagging behind its international peers who have established centers and/or collaborative facilities that focus on advancing HEA research and application. Examples of international efforts include: (1) the Center for High Entropy Alloys, Postech, Korea; (2) the Danish Center for High Entropy Alloy Catalysis; (3) High-Entropy Alloys Research Center, State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing; (4) the TURBO-AHEAD project in France; and (5) the German DFG Priority Programme on Compositionally Complex Alloys. Furthermore, it is not clear how existing US-based national materials development efforts, such as the more broad-based Materials Genome Initiative (MGI), can best support novel materials discovery, development, and deployment approaches required specifically for HEAs. It is important, therefore, that the US establish a national network of HEA interconnected partnerships (e.g., Centers of Excellence) to maintain leadership in this revolutionary technology.

Developing any nationally coordinated group of centers in materials science comes with a characteristic set of challenges and impediments. Novel materials often require new or modified equipment and infrastructure to be designed and developed (e.g., see Action Plans 1 and 2). Transitioning from laboratory results to commercially available products often requires additional research, since the synthesis and thermo-mechanical processes used for small samples are different from those used at the industrial scale. Due to the potential profitability of impactful, commercially viable HEAs, network partners will surely raise intellectual property (IP) issues related to protection and classification of developed HEA materials and datasets. While these challenges are not unique to HEAs, they may be more substantial than for conventional alloys.

The underlying reason for this is that HEAs are not so much a new class of alloys with distinct but limited processing approaches and application potential, but rather represent a new approach for designing inorganic materials. As a result, HEAs represent a much broader range of processes and applications than any single alloy class, and the extent of these challenges may be accordingly broader.

Development of a nationwide network of interconnected HEA partnerships can go a long way toward helping overcome many of these challenges, and the tasks outlined below provide some detailed recommendations of how this might be accomplished.

Task 3.1–Form a government agency working group, team, or oversight committee

A first step in developing a national HEA research effort in the US is to convene a group of stakeholders among the various federal agencies to discuss such an initiative. Representatives from various government agencies, such as the Department of Defense (Army, Navy, Air Force, and DARPA), DoE, NSF, National Aeronautics and Space Administration (NASA), and Department of Commerce (NIST) could be invited during the formation of this committee. Initial milestones would be to convene a kickoff meeting and schedule regular subsequent meetings (i.e., quarterly or semi-annually) to build consensus and avoid unproductive overlap on HEA topical areas, and to discuss possible pathways to establish a national network of interconnected HEA partnerships, focused on HEA topics. This activity has the distinct advantage of being achievable within a year's time frame and with minimal associated costs.

Task 3.2–Establish a national network of HEA Centers of Excellence (CoEs)

To rapidly and effectively advance widespread HEA research and development, it is recommended that a national network of interconnected HEA Centers of Excellence (CoEs) be established, in which each CoE focuses on distinct yet complementary topics critical to the HEA ecosystem. For example, CoE activities could include (1) high-throughput/autonomous research (see Action Plan 1), (2) modeling, theory, and data acquisition strategies, (3) HEA-specific user testing facilities, and/or (4) the exploration and development of HEAs for specific application domains. A range of functional and structural application domains is recommended to ensure the full potential of the HEA approach is achieved. To ensure that each CoE has the comprehensive capabilities needed to accelerate discovery and development for its chosen specialty or application class, strong partnerships between university, industry, and government laboratories are expected for all CoEs. A CoE might typically be university led, but since government laboratories often address specialized or unique challenges and applications, selected CoEs may also be led by government laboratories. Regional centers dispersed across the US would ensure that the full spectrum of innovation available at universities and at small and large businesses, including global companies with a regional presence, are engaged in a given CoE. This national network of regional CoEs also enables a diverse workforce to be trained in and contribute to this developing technology. These CoEs will benefit the US by developing the methods, materials, and associated technologies needed to establish new domestic capabilities for defense applications, as well as for high performance export industries such as aerospace, automotive, energy, and medical industries.

This national network of interconnected, regional CoEs could look to existing multi-center initiatives, similar for instance to the DoE's Energy Materials Network,[209] for guidance on integrating multiple research and/or technology hubs into a coordinated effort to expedite the discovery, development, and deployment of a particular class of material. Such existing entities exemplify the advantages of a multi-stakeholder partnership approach, which takes advantage of coordination amongst industry, national laboratories, and universities to capitalize on unique capabilities and application spaces.

Initial milestones for this activity are to determine: (1) various center-level potential topics of interest for HEAs, (2) the desired number of CoEs, and (3) the size and scope of each individual CoE. The identification of these three criteria should be spearheaded by the committee recommended in Task 3.1 and could take 1–2 years to settle on a final consensus. Once funded, each CoE might receive on the order of $10M annually for at least 5 years, from a primary funding source, in order to see meaningful gains.

Task 3.3–Develop strong academic–industry–Government partnerships for HEAs

As discussed in Action Plan 1 and in Task 3.2, significant involvement and collaboration between academia and industrial stakeholders is required to unlock the full potential of HEAs. As academics develop new theories necessary to guide the exploration of promising alloy systems and applications, industrial partners are needed to develop specialized equipment to enable laboratory- and commercial-scale HEA fabrication and testing, and ultimately to transition the science and technology to products. Each of the CoEs described in Task 3.2 should thus include industrial, academic, and government partners. The work of these groups would be significantly improved through regular communication, collaborations, and, in certain instances, the founding of novel businesses to satisfy demands unique to the HEA space. Therefore, another foundational activity for HEA development is to strengthen small business capability. A key, initial milestone for success in this area is for interested agencies to map out and promote a plan to leverage existing SBIR programs to expedite the advancement of multi-principal element alloys and their enabling technologies. The initial outlining of how best to promote the need for strong academic–industry partnerships in HEA development can be done in conjunction with Task 3.1 and 3.2 above. Subsequent SBIR support on the order of $5M per year over the next 4–5 years could be dispersed to a plethora of collaborative teams in order to securely establish a close relationship between academia and industry that will only grow stronger as this field evolves.

Task 3.4–Launch a DoD-led consortium on high-throughput/autonomous research of HEAs

The development of a high-throughput, autonomous research infrastructure is foundational to the success of any program geared to effectively finding and refining impactful high entropy materials in the future (see Action Plan 1 for details). Therefore, it is important that a consortium is created which serves as a coordination hub for the various development activities around high-throughput screening, modelling, synthesis, and experimentation involving HEAs. In the spirit of the NSF Big Data Hubs for instance,[210] this consortium could grant awards to investigators performing research on high-throughput capabilities applicable to HEAs of interest to Department of Defense (DoD) applications. In addition, other established industry-based consortia, such as the Metals Affordability Initiative (MAI) of the Air Force Research Laboratory (AFRL),[211] can be leveraged to affordably test, produce, and implement attractive HEAs. A DoD-led body focused on high-throughput, autonomous research of HEAs would be ideally positioned to: (a) provide overarching direction in this currently disjointed, emerging field, (b) promote joint collaboration and publications, and (c) ensure knowledge sharing across interconnected yet often siloed research activities. Some metrics of success for this group could include: (1) the number of formal grant proposals which receive an award, and (2) the number of joint publications and patents that come from said awards. To construct a comprehensive consortium such as this would likely take 3 years and perhaps $15M for an initial wave of funding. This task also relates to the high-throughput CoE discussed in Task 3.2.

Task 3.5–Assign specific HEA topics to future CoEs

As discussed in Task 3.2, a subsection of particularly intriguing HEA related topics should be identified for CoE-level engagement. Example specialties may include (but not be limited to) specific production/processing methods, characterization, testing, and/or application classes. Examples of possible production/processing methods include additive manufacturing, high-energy ball milling, powder atomization, and/or ingot manufacturing. Characterization and testing methods are considered in Action Plan 1 and in Section VI of this report, while Section IV discusses some target application areas. There can be latitude for a great amount of flexibility between the structure of the various CoEs. For example, each CoE may focus their activities on projects at specific technology readiness level (TRL) ranges, and/or within certain scientific or technological domains. In any case, some CoE success metrics that may be applicable include the number of new materials which have been delivered/distributed for investigation across the CoE network, as well as the number of papers, citations, patents, and/or patent disclosures generated from the work. A given CoE might require $5–10M annually.

Task 3.6–Launch a HEA CoE for theory, modeling, and data acquisition/management strategies

The necessary steps to develop new fundamental theories, informative computational models, and appropriate data acquisition strategies have been outlined in detail in Action Plan 2. Any HEA CoE coordinating these activities may leverage existing materials-related data and/or modeling centers (e.g., ChiMaD, nanoHUB, PRISMS/Materials Commons). This particular HEA CoE would require leaders and participants with various types of specific expertise, including physicists, materials scientists/engineers, applied mathematicians, and a diverse team of data scientists, engineers, and analysts. This would likely include a mix of people with representation from academia, national laboratories, and industry. Moreover, various federal agencies and national laboratories should be approached to provide access to High Performance Computing (HPC) centers and resources. Significant participation from these groups of stakeholders, as well as metrics such as resultant number of publications and citations, are some ways to measure the success of this venture. Equally important to developing validated models will be making these models widely available to domain experts (beyond the computational materials science community) who can use these models to speed up the design and fielding of new materials. Establishment of this CoE is expected to require on the order of $3–4M per year, for three years, depending on the level of engagement and equipment support needed. The cost to continue to run such a center should drop significantly after the three year start-up period.

Task 3.7–Launch a HEA CoE for technology transfer and scale-up

Once a HEA material has repeatedly demonstrated promising performance at a laboratory- scale, there needs to be an established pipeline to expedite the technology transfer and scale-up process, in order to enable rapid deployment into DoD applications and commercial markets. This CoE would essentially function as a bridge, pilot-scale manufacturing center, including some major milestones associated with the tasks outlined in Action Plan 4 below. Some key metrics to measure and document for this center are: (1) the demonstrated materials performance, and (2) any relevant compositional, processing, microstructural, and testing data which can inform large scale production. Establishing a CoE specializing in technology transfer and scale-up would require significant industrial input at various stages, concerning the development of mid- and large-scale testing equipment, real-world testing opportunities, and a high level of understanding of market and industry trends, as well as Intellectual Property (IP) law and resources. Examples of potential industry partners include ATI Specialty Alloys & Components, H.C. Starck, Carpenter Technology: Specialty Alloys, Alfa Aesar, and Retech Systems (SECO/WARWICK). In addition to a strong industrial presence, this CoE should have significant representation from various branches of the DoD, to advise on relevant national security applications. Finally, this center should also have access to legal counsel that can advise on various patent and commercialization issues. It is expected that establishment of this CoE would require as much as $3M per year for up to 3 years.

Task 3.8–Launch a HEA CoE for design-quality test data collection, storage, analysis, and sharing

As more full-scale HEA CoEs become operational, there will be untold amounts of data output from their various activities. In order to corral this mountain of data into meaningful, actionable progress, it is critical that a FAIR-principled[202] repository for design-quality test data be established, in which data from all the HEA partnerships and/or centers is stored and organized. This repository should be accessible by all consortium members to help enable leveraging, accelerate development and design of new HEAs, and provide continuity to development and design approaches across the network's partnerships and/or CoEs. An important objective of this particular CoE's leadership will be to create a data infrastructure that manages the disparate data types needed for MPE alloy development, including metadata, for the aforementioned repository. This repository should not only effectively catalogue and correlate relevant HEA design and development data but can serve as a model schema for other data repositories housed at partner CoEs as well (see Task 3.2, and Action Plan 2, Task 2.6 as well). Creating this CoE would require insight from national laboratories, industry, universities, and certified test laboratories, as well as the technical expertise of data scientists, computer scientists, and engineers. Once operational, some metrics to measure progress could include the amount of data aggregated, the number of people supplying data, and the number of people accessing the data. Technology transfer-related metrics could include, for instance, the number of design curves generated, and how many of them yielded promising final products. It is expected that establishing this CoE could require on the order of $25M spread over a period of 5 years or so.

Action Plan 4: Engage in Feedstock Production and Scalability Research

This action plan is primarily related to challenge areas A—*High-Throughput Screening Methods and Experimental Tools*, and H—*Availability of Affordable Powder*, from Section V. It dovetails best to the following preliminary recommendations (from Section VI):

- Develop a high-throughput processing approach for bulk HEA samples (A7)
- Establish a research-scale powder manufacturing facility for HEA development (H32)

The motivation for this action plan is the challenge of obtaining the required quantity, quality, and size of HEA specimens needed for HEA research, development, and scale-up efforts. For example, MPE alloys have unique processing needs (especially RHEAs). In this vein, it can be difficult to achieve high levels of quality and throughput in the production and processing of refractory materials, due to effects related to their high melting temperature. Additive manufacturing (AM) is a promising production route with great potential for HEAs, yet it does present some challenges. For instance, additively manufactured samples can differ greatly from alloys that are produced using traditional solidification, and there is often insufficient understanding of AM process fundamentals with respect to HEA feedstock volume and control. The current methods for production of refractory powders are strongly challenged by alloys requiring four or more refractory elements with a minimal content of interstitial elements. Finally, partner facilities (i.e., authorized powder production facilities) may raise IP issues related to open data sharing for legacy and emerging HEA feedstock materials. The recommended tasks below will help to address many of these issues.

Task 4.1–Establish a small-batch production facility

This facility would provide small-scale experimental heats, and/or batches of powder, for research purposes. Such a facility would probably best be established (and maintained) by a commercial vendor with knowledge and experience in this domain. This vendor might also have a vested business interest in such a new product stream. This will help share the cost and risk of the venture, provide further motivation for achieving the capability, and could also accelerate technology transition. Perhaps some existing powder supplier or foundry shop could fit such a role. This task will require dedicated processing facilities for specific powders and/or heats, and establishment of such a capability could take anywhere from 1–3 years. Although there is some uncertainty regarding the required cost of labor and other details, a ballpark estimate for the cost to develop such a facility would be on the order of $2–4M. It should be a shared facility that becomes accessible to a broad community of researchers who can acquire starting powders and/or small ingots of desired HEA compositions. A metric of success would be how many researchers take advantage of this resource and acquire such materials for their research. Another success metric would be the longevity and fiscal stability of the facility. Companies that might establish such a capability could include new and/or established small businesses. National laboratories could partner with these companies, providing both expertise and/or some start-up support (at least in-kind) to help get such a facility off the ground.

Task 4.2–Identify existing powder production facility partners to aggregate feedstock demand

The idea here is to seek out existing facilities that can provide a service to the field, but do not necessarily see themselves as a direct part of the research infrastructure. Some specific examples of the types of industry partners that might be involved include companies like ATI Specialty Alloys & Components, H.C. Starck, and Alfa Aesar, since existing equipment at these companies can be adapted for HEAs. A consortium or some alternative mechanism could be established to aggregate demand from individual groups involved in HEA research and/or technology transition in order to more easily obtain HEA feedstock, including scale-up volumes of material. This task thus dovetails with Action Plan 3 on developing a nationwide network of interconnected HEA partnerships. One mechanism to join the consortium proposed in this task could include "user proposals" in which justification for a particular research and/or development team is provided. Establishment of this feedstock production network and related equipment could take 2–3 years, with year 1 including the hiring of personnel with expertise to operate equipment for the consortium. Metrics of success could include the contribution of this consortium to the technical goals of externally funded projects, the number of publications resulting from those projects, and the resultant impact of those projects on the development and/or deployment of new HEA materials. National laboratories could also partner with such a consortium. The estimated cost to establish such a consortium is $5M annually during the establishment period of the consortium (first 1–2 years).

Task 4.3–Introduce provenance requirements for HEA databases

This task dovetails with Task 3.8 and other data-related tasks. As a robust HEA database accessible to a wide variety of researchers and engineers is developed, it is critical to provide the proper provenance of the materials, experimental conditions for testing and characterization data, and boundary conditions and limitations for computational data. Data provenance can be defined as "…metadata that is paired with records that details the origin, changes to, and details supporting the confidence or validity of data."[212] Without such provenance, researchers and developers will not be able to responsibly use such data. In other words, this is a mechanism of establishing "ground truth" within HEA materials and properties databases.

Under this task, facilities and/or database leaders or curators should require provenance of all HEA materials and/or properties data from all data providers, as well as provide such metadata to all data users. This activity should be initiated immediately and may take up to three years to fully implement (including for existing databases). The metadata could also be analyzed and help support the community in establishing uniform metrics for purity and processing of HEA materials (e.g., tighter parameter spaces for alloy purity, temper treatments, etc.). The cost of this task could be on the order of $1M annually for a period of 1–3 years.

Task 4.4–Provide researchers access to thermomechanical processing facilities

Access to thermomechanical processing facilities for HEAs will accelerate efforts toward the development of HEA materials and components for application. For instance, researchers at government agencies and universities, and their associated research partners, could be granted access to facilities for thermomechanical processing at government laboratories and/or within HEA consortia (e.g., see Task 4.2 and Action Plan 3). A proposal system could be devised in order to ensure access by high-quality research programs, as well as those with the most critical need.

These facilities would ensure more uniformity and reproducibility of specimens and/or their properties. A metric of success could be the total number of customers served annually by these resources. Providers of these facilities could include federal agencies, national laboratories (e.g., the Air Force Research Laboratory (AFRL) or Oak Ridge National Laboratory (ORNL)), industrial partners in HEA consortia (see Action Plan 3), and/or small businesses. This activity could begin almost immediately for existing facilities, and perhaps take up to three years to bring other facilities on board, retrofit some existing equipment, and/or build/purchase some new equipment. Costs for this task could range from $500,000 per year for maintenance of existing equipment in consortia, to $5M or more to help assemble new facilities.

Task 4.5–Research new powder production methods

Under this activity, researchers in academia, government laboratories, and/or industry would conduct research on new powder production methods for HEAs. These efforts could include assessments of potential scale-up volumes, relative to current approaches, as well as novel new methods of laboratory scale production. They would also involve characterization of the chemistry and quality of powders. Costs associated with this activity are estimated to be on the order of $150,000 annually for two research groups/PIs, over 3–5 years (i.e., anywhere from about $1M to $1.5M).

Task 4.6–Establish high-throughput screening protocols to assess HEA processibility

This task dovetails directly with Action Plan 1. High-throughput screening protocols are needed to assess the processibility of emerging HEAs. Researchers at universities and national laboratories can demonstrate these protocols on known materials, and then extend them to emerging materials, in coordination with industrial collaborators. A metric of success is how many industrial companies eventually pick up and use these screening protocols and/or the screened results, in their efforts to develop new components or product lines based on HEAs. Costs associated with development of these high-throughput screening protocols to assess the processibility of HEA materials would be on the order of $1–2M; that is, $150,000 annually for two or three research groups, over 3–5 years.

Task 4.7–Generate processing and property data for legacy and emerging HEA materials

This activity involves generating robust processing and property data on legacy and emerging materials, particularly to develop large HEA datasets amenable to machine learning techniques. This task dovetails with Tasks 2.6 and 2.7 in the computational realm (Action Plan 2), but here the focus is on experimental data. For associated databases developed outside universities, a plan would have to be developed for database maintenance as well. This activity would involve multiple research teams developing data over a wide range of compositions, processing conditions, and properties. These teams could be based at universities, national laboratories, and/or industrial companies. Although IP issues might restrict the data that industrial companies could share, there may be some pre-proprietary types of data that could be considered of mutual benefit for inclusion in public and/or consortia databases. A rough cost estimate for generation of this type of robust data might thus be 7 research teams/PIs at a cost of $150,000 annually each, for 3–5 years. This corresponds to a total cost in the range of about $3–5M, but much of this cost could be shared with the aforementioned related tasks. Costs for any database maintenance would be over and above these estimated costs. Small and medium-size companies working in the area of materials data (such as Citrine Informatics, Ansys/GRANTA, Materials Data Management, etc.) could also be important contributors to such efforts.

Task 4.8–Develop melt-less processes for ultrahigh-temperature materials

HEA candidate materials and components for ultrahigh-temperature applications are very difficult to produce by processes exceeding their melt temperatures. This task would encompass the investigation and development of melt-less processes. These could include powder consolidation via some combination of sintering and/or hot isostatic pressing (HIPing), and/or some other type of thermomechanical processing. Eventually a pilot scale facility(ies) would have to be built to conduct assessment of feasibility for production at relevant volume scales. Such pilot facilities might not address full industrial scale-up but would be a steppingstone from laboratory specimens to material volumes at the next level of scale. These research and development efforts coupled with the need for pilot scale-up facilities may cost between $10–20M and take up to 5 years. They would involve a combination of researchers at universities and national laboratories, as well as scientists, engineers, and/or designers at both large industrial companies and smaller businesses.

Task 4.9–Conduct research on powder recycling issues

Under this activity, protocols would be developed for reuse and recycling of powders used as HEA feedstock. These protocols would most likely be developed by scientists and engineers at universities, or government laboratories, within a timeframe of about 2–4 years, and costs between $500,000–$1M annually.

Action Plan 5: Develop Training and Resources for Workforce Readiness

This action plan is related to challenge area I in Section V - *Workforce Trained in HEA Exploration and/or Development*. It dovetails with the following preliminary recommendations from Section VI:

- Establish graduate-level internships and cooperative educational programs (I34)
- Produce a multi-institutional workshop series (I36)

Specific challenges and opportunities that provide the motivation for this action plan include the current lack of undergraduate and graduate curricular materials on non-dilute multi-component materials topics. Along these lines, there are few venues to raise awareness and train potential industry end-users or practitioners in the scientific underpinning, engineering aspects, and utility of HEAs. There are also limited university laboratory experiences to prepare undergraduate or graduate students for experimental methodologies that are especially needed for the discovery, development, and implementation of HEAs. In particular, laboratory modules and/or design projects that engage high-throughput, automated experiments are critical.

Task 5.1–Organize courses and/or workshops on applicable computational techniques

Curricula on computational techniques applicable to HEAs should be developed for undergraduate students, graduate students, and/or professionals (i.e., for continuing education). Due to the barriers and difficulties associated with inserting new courses into an already packed curriculum in most university departments (see also task 5.3), initially courses could be provided as summer courses at universities or short courses offered by professional societies.

In a related fashion, workshops on computational tools for HEAs geared toward educators (teaching assistants and instructors) can be developed, i.e., to "educate the educators". Similar workshops on lab-based activities in HEAs could also be developed. These summer courses and/or workshops could be offered by universities with large existing MSE (or related) departments, and/or relevant professional societies. A rough estimate of cost associated with developing and executing such short courses and/or workshops would be on the order of $50,000 to $70,000 per offering, but much if not all of that cost would be recouped in tuition/registration fees. One metric of success would be the total number of students and industry professionals attending these offerings. The time frame for commissioning these activities would be relatively fast for professional society short courses and/or workshops (1–2 years) but would likely be longer for summer courses within university departments (perhaps 2–4 years).

Task 5.2–Identify internship opportunities

A complimentary activity to Task 5.1 for creating excitement in and educating the future workforce in HEA discovery, development, and implementation is to create associated undergraduate and/or graduate-level internships. These could be developed in coordination with university cooperative education offices to identify topics and partner institutions within industry or government laboratories that have HEA-relevant interests, capabilities, and personnel. Specific role players for these internships would be university departments with existing internship programs and/or perhaps those that are NSF Research Experiences for Undergraduates (REU) sites. Industry partners and/or government laboratories with relevant HEA interests and capabilities would be critical to serve as hosts for these internships. In the government laboratories, initial focus could be on DoD laboratories that already have research programs in the HEA arena. Estimated costs are on the order of $10,000 per internship. Especially by leveraging existing infrastructures, HEA-related internships could be started within the next year or two. Metrics of success for this task would include the number of commitments secured from industry and government laboratories to host such interns, and subsequently the number of internships taken by students.

Task 5.3–Incorporate HEA topical foci into Materials Science & Engineering (MSE) curricula

This task is centered around creating new modules, or modifying existing MSE curricula, to incorporate HEA topics. Ready-to-use learning modules should be created, including lectures, homework assignments, and laboratory sessions. Existing MSE courses could be modified or new MSE curricula developed, in order to address HEA topics such as the large number of alloy degrees of freedom, cocktail effects, and methods for addressing structural and chemical complexities. Undergraduate HEA modules at both introductory and/or more advanced levels should be developed on topics including (but not limited to) mechanical properties, corrosion, thermodynamics, and phase diagrams related to HEAs. HEA-related computational modeling curricula for graduate-level students is also recommended. Changing curriculum will likely require weigh-in from each state's Board of Regents.

A survey could be disseminated to industry, national laboratories, and start-up companies on current workforce capabilities and skills needed to help develop content crucial to educating the future workforce. In this regard, multi-institutional partnerships could be very beneficial for this activity, as all these entities can provide critical input on current workforce capabilities and skills needs.

Adjunct professors could be invited from government laboratories to teach and/or play a role on thesis committees. Consideration should also be given to obtaining seed grants or start-up support for faculty-industry-national laboratory partnerships.

All of these activities could dovetail with educational workshops and summer courses (Task 5.1), as well as HEA-based symposia, with the goal of sharing resources geared toward course content. Collaborative opportunities between materials science and engineering (MSE) and data science educators/programs should be explored, including development of materials-focused homework, course content, and/or project topics for data science courses, and joint advisership on graduate theses.

Estimated costs are about $50,000 for each module developed (e.g., for a given semester), and these offerings should be able to be provided within two to three years. Measures of success would include the number of modules offered across various universities and departments, and the number of students that attend these modules. ABET-type (i.e., Accreditation Board for Engineering and Technology) assessments of outcomes could also be conducted, as well as student surveys, to explore the degree of success of these modules. Academia should certainly engage and leverage the ABET infrastructure in this regard.

Task 5.4–Develop HEA textbooks and/or chapters

New reference textbooks on HEAs, or HEA chapters within broader books, are critical to the education of the HEA workforce. These should be offered in electronic format as well as in print. Texts at both undergraduate and graduate levels should be developed. Content would be related to non-dilute, multicomponent materials, and span many learning domains, including thermodynamics and alloy development, kinetics (diffusion), phases and microstructure, properties, and characterization methods, among others. HEA book chapters could be created and inserted into "standard" core undergraduate textbooks. A series of homework problems could be included at the end of these chapters. Researchers in academia who write these chapters, or full textbooks, could engage publishers, and/or professional societies who have strong relationships with publishers. The costs should be absorbed by the revenue that the books produce. Book chapters could be written and published within the next 1–3 years, whereas full books might take a longer time horizon for publication.

Task 5.5–Create certificates and/or credentials associated with short courses

This activity would encompass specialized programs focused on the role of HEA-related practitioners and/or technicians, and related certificates or credentials for those who complete the course/program and would thus dovetail with Task 5.1. Professional societies would likely have a role in developing specialized programs with related certificates or credentials. Some programs might provide credits that could be applied toward an associate degree, or if done through a university, perhaps even a bachelor's degree. Although there will be some start-up costs to develop materials, other programs or short courses of this nature have previously delivered a full return on investment, based on the revenue from the registration fees. These programs could be developed within 2 or 3 years.

VIII.

Closing Remarks

High Entropy Alloys (HEAs) offer promising combinations of properties and revolutionary potential for superior materials and component performance, particularly for defense applications. HEAs were conceived less than 20 years ago, and they cover a vast array of compositions across a wide alloy space, so efforts still emphasize relatively fundamental aspects of scientific research. As a result, the development of clear pathways toward implementation of HEAs for specific applications is in its infancy. HEAs are not ready at present to produce products at scale. High entropy brasses and bronzes, described in Section IV, are perhaps an exception and may be closest to transitioning to application.

This science and technology accelerator study report on *Defining Pathways for Realizing the Revolutionary Potential of High Entropy Alloys* consolidates information available in the literature with the ideas and recommendations of a group of internationally recognized experts from academia, industry, and government working on research and development of HEAs. The strong value proposition for HEAs (Section III), challenges and needs (Section IV), target application areas (Section V), preliminary recommendations (Section VI), and in-depth action plans and recommended tasks (Section VII) are all provided to help the research and engineering community make great strides with HEAs, particularly in support of national security interests, but also for broader economic and societal benefits. Such strides include science and technology breakthroughs that support realization of the wide-reaching potential of HEAs in key, prioritized areas, in order to accelerate the discovery, development, and eventual use of these potentially disruptive materials.

Scientists, engineers, systems and component designers, technical leaders, those in a position to provide financial support, and others who read this report can use the information here to stimulate direct action. Readers can act upon the recommendations, action plans, and detailed tasks presented in this report almost immediately. These recommendations, action plans, and tasks are not all-inclusive, and readers of this report can build on this information to stimulate the development of additional ideas and activities that may further contribute to the development and implementation of HEAs.

It is our hope that the content in this report is informative, and that it inspires new efforts to advance the field of HEAs by addressing the action plans and recommended tasks outlined here. This can be done by initiating and contributing to new technical efforts within your organizations or by providing support and/or guidance for such activities. There is great potential to accelerate the development and implementation of these potentially revolutionary materials, in order to help produce what could be a new generation of advanced materials and products of great benefit for national security, the economy, and society as a whole. The time to act is now.

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Appendix: Glossary

Complex concentrated alloys (CCAs)	Includes all alloys in the HEA field, as well as alloys that satisfy the motivation of studying complex, concentrated alloys or the vast number of compositions and microstructures in the central regions of multi-component phase diagrams
FAIR Guiding Principles for scientific data management and stewardship	Findability, Accessibility, Interoperability, and Reusability
High Entropy Alloys (HEAs) and High Entropy Ceramics (HECs)	Metallics, ceramics, metal-ceramic composites; single- and multi-phase solid solutions; 5 or more principal elements, each molar ratio 5-35%; not exclusive to equiatomic HEAs
High- and ultrahigh-temperature thresholds	High-temperature: $T \geq 1300°C$ Ultrahigh-temperature: $T \geq 2000°C$
High-throughput experiments	The acceleration of experimentation through combinatorial methods and/or automation such that high rates become feasible
Larson-Miller Parameter	The most extensively used extrapolation parameter for predicting creep life of metallic materials
Light Water Reactor	A type of thermal-neutron reactor that uses normal water, as opposed to heavy water, as both its coolant and neutron moderator
Medium-Entropy Alloys	2-4 principal elements; entropy between R and 1.5R

Molecular Dynamics (MD)	A computer simulation method for analyzing the physical movements of atoms and molecules, allowing interaction for a fixed period of time in order to give a view of dynamic "evolution"
Monte Carlo	A broad class of computational algorithms that rely on repeated random sampling to obtain numerical results
Multi-Principal Element (MPE) Alloys	A new alloy development philosophy, where the base alloy has significant atom fractions of several elements. *See* Complex concentrated alloys (CCAs)
Reduced Order Model (ROM)	Simplifications of high-fidelity, complex models that can be used by scientists and engineers to quickly study a system's dominant effects using minimal computational resources.[195] ROMs sacrifice some accuracy and robustness for speed.
Refractory High Entropy Alloys (RHEAs)	HEAs with relatively high melting points (e.g., 1800°C) intended for ultrahigh-temperature structural applications.
Thermal Barrier Coating	An advanced materials system usually applied to metallic surfaces operating at elevated temperature (such as gas turbine or aero-engine parts)
Uncertainty Quantification	The process of quantifying uncertainties associated with model calculations of physical quantities of interest (QOIs), with the goals of accounting for all sources of uncertainty and quantifying the contributions of the specific sources to the overall uncertainty,[198] due to inherent variability (irreducible uncertainty) or lack of knowledge (reducible uncertainty).
Verification and Validation	Verification: the process of determining that a computational model accurately represents the underlying mathematical model and its solution.[197,198] Validation: the process of determining the degree to which a model is an accurate representation of the real world from the perspective of the intended uses of the model.[197,198]

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