High-Entropy Alloys

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FOREWORD

In the 1970s, I became excited about the idea of multicomponent alloys. I realized that the materials we use are almost all based on a single component with a primary property and an admixture of small amounts of other components to provide secondary properties. Effectively, all our known materials are at the corners and edges of a multicomponent phase diagram consisting of all possible components. This means that there is a vast number of possible materials in the middle of this phase diagram which have never been investigated. It turns out that this unknown field of materials is truly enormous, and the number of unexplored materials is many times greater than the number of atoms in the universe.

This explains why we keep discovering exciting new materials: high-temperature superconductors, glassy alloys, quasicrystals, compound semi-conductors, and so on. And we will keep finding exciting new materials as long as we have the courage as experimenters to try innovative, new mixtures of constituents. I now tell every PhD student in materials science to be aggressive and ambitious in exploring this amazing array of potential new materials.

In the 1970s, I found it hard to persuade other people to be similarly enthused by my ideas on the topic of multicomponent alloys. I could not get funding, and I could not get research colleagues to undertake preliminary experiments. Everyone wanted to work in much better known fields. Everyone was very conservative. Finally, I persuaded a young undergraduate student, Alan Vincent, to do preliminary work. He immediately found exciting results including the first high-entropy alloys. Nearly 20 years later, I persuaded another young undergraduate student, Peter Knight, to repeat the work. And finally, a couple of years later, my long-standing research colleague, Isaac Chang, repeated the experiments for a third time, but more carefully and with more time to document the results fully and in a publishable way. We presented the results, first found by Alan Vincent in 1979, at a conference in 2002 which was published in 2004, more than 25 years since my first idea. In parallel, Professor S. Ranganathan (my old friend, Rangu) and Professor J.W. Yeh (my new friend, Jien-Wei) published independently papers on, respectively, material cocktails and high-entropy alloys, closely related and essentially similar concepts to my idea of multicomponent alloys.

In the last few years, as a consequence of the outstanding continuing work by Jien-Wei, the field of multicomponent and high-entropy alloys has taken off, with literally hundreds of publications each year. Most notably, Vincent, Knight, Chang, and I discovered in the late 1970s a single FCC solid solution consisting of five components in equal proportions, namely, FeCrMnNiCo. This alloy has been shown to have outstanding mechanical properties, with high strength and high ductility. I realized in the late 1970s that the mechanical behavior of this material would be very unusual. Metal and alloy mechanical properties depend primarily on the behavior of dislocations and how they move in response to stress, but the concept of a dislocation as a line defect with a consistent core structure becomes complex when there are many different components distributed on a single lattice.

Professors Murty, Yeh, and Ranganathan have now written a book on this new group of materials. The book covers the structure, processing, and properties of the materials, insofar as we have been able to explore them. Some multicomponent alloys are solid solutions with high entropy. Some are not. In either case, there are wonderfully exciting new structures and properties to be found. This book is the first about this field. It contains many valuable and interesting insights. But ultimately it can only hint at the full range of new materials which remain to be discovered. The authors should be congratulated on doing an important job which will help us on our exciting, exploratory journey into the materials of the future.

Prof. Brian Cantor
Vice Chancellor,
The University of Bradford, UK

PREFACE

Alloys traditionally have been based on a solvent element to which various solute atoms are added for improving specific properties. Thus, alloys are usually named after the major element in the alloy (e.g., Fe-, Al-, Cu-, Mg-, and Ni-base alloys). Two people, in recent times, have changed the way people look at alloys and they are Prof. Brian Cantor and Prof. Jien-Wei Yeh by coming up with equiatomic and nonequiatomic multicomponent alloys. Incidentally, though each of them started working on these alloys independently at different times (Cantor starting in 1979 and Yeh starting 1996), their work came to open literature in the same year, 2004. Interestingly, even before the papers of these two pioneers got published in 2004, Prof. S. Ranganathan felt the importance of this new class of alloys and wrote about them in his classic paper "Alloyed Pleasures: Multimetallic Cocktails" in 2003, which has been cited more than 100 times now as the first publication on this class of alloys.

Yeh christened these alloys as "high-entropy alloys (HEAs)," rightly so, as the configurational entropy of these alloys is expected to be very high at their random solution states. Such a high entropy is expected to drive the tendency to form simple solid solutions (crystalline or amorphous) rather than complex microstructures with many compounds. The concept has caught the attention of many researchers and the last one decade witnessed about 400–500 papers being published on HEAs with various elemental combinations. Two major observations can be made from all this work, namely, the alloys do form simple solid solutions in most of the cases and the number of phases observed in these alloys is much less than the maximum predicted from the Gibbs phase rule.

There are also clear indications that the high entropy in these multicomponent equiatomic and nonequiatomic alloys is not able to act like a glue holding all the atoms together in a single solid solution, and there are reports on the formation of two or more phases in which intermetallic phase formation and even segregation of certain elements were observed. This could be related with the various thermodynamic and kinetic factors. There has been intense activity in past few years to predict the phases that can form in such multicomponent alloys through various modeling approaches including integrated computational materials engineering (ICME) using various tools such as CALPHAD, *ab-initio*, molecular dynamics, Monte Carlo, and phase field approaches, which have been supported by materials genome initiative (MGI).

Besides the scientific curiosity, researchers also feel that HEAs can substitute conventional materials in advanced applications so that the limitations of the latter in service life and operational conditions could be overcome by providing superior performance of the former. A number of processing routes, including conventional melting and casting, mechanical alloying, various coating techniques, and even combinatorial materials science approaches are being used to synthesize and process this new class of alloys. There have been a lot of studies on understanding both the structural and functional properties of these alloys. The results of HEAs and HEA-related materials reported so far by various research groups are very encouraging for their applications in a wide range of fields such as materials for engine, nuclear plant, chemical plant, marine structure, tool, mold, hard facing, and functional coatings.

It is just over a hundred years since Walter Rosenhain wrote his seminal book on Physical Metallurgy. A century of research resulted in spectacular progress but the field became mature and the excitement began to wane. At the beginning of the third millennium, the discovery of High-Entropy Alloys has ushered in a renaissance in physical metallurgy.

This book presents a comprehensive insight into all the above aspects of this exciting new class of alloys. The book, being a short format one, is written keeping a beginner in the field in mind to give him/her an idea on various facets of HEAs that he/she can pursue.

B.S. Murty, J.W. Yeh and S. Ranganathan

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The authors are grateful to Prof. Brian Cantor for readily agreeing to write a foreword for the book, in spite of his hectic schedule as Vice Chancellor of the University of Bradford, UK. They are also highly indebted to all the authors of various papers that are referred to in this book. Their contributions have enhanced the quality of the book.

Prof. Murty would like to specially thank his group members, Mayur, Guruvidyathri, Anirudh, Arul, Ameey and Dr. Sanjay, who have untiringly helped at various stages of the book. He would also like to gratefully acknowledge the collaboration with his students, Varalakshmi, Praveen, Pradeep, Sriharitha, Ashok, Durga, Raghavan and collaborators Dr. Ravi Sankar Kottada, Prof. M. Kamaraj and Prof. K.C. Hari Kumar, Dr. Sheela Singh, Dr. N. Wanderka and Prof. J. Banhart and Prof. D. Raabe. He is also indebted to his family members for their patience and continuous support.

Prof. Yeh would like to thank two important group members, Prof. Su-Jien Lin and Prof. Swe-Kai Chen, for their long-term contributions in the research on high-entropy alloys since 1995. He also expresses sincere thanks to Profs. Tsung-Shune Ch in, Jan-Yiaw Gan, Tao-Tsung Shun, Chun-Huei Tsau, Shou-Yi Chang, Tung Hsu, Wen-Kuang Hsu and all graduate students for their significant efforts and contributions in HEA-related research. In addition, he specially thanks Dr. Chun-Ming Lin and PhD student Chien-Chang Juan for their help in searching related papers, plotting figures and providing suggestions for this book. Finally, he gratefully thanks his beloved wife and daughters for all their encouragement and support during writing this book.

Prof. Ranganathan records his gratitude to numerous teachers, colleagues and students who instilled in him an abiding interest in physical metallurgy. Special thanks are due to T.R. Anantharaman, K.P. Abraham, C.N.R. Rao, P. Rama Rao, Alan Cottrell, David Brandon, Robert Cahn, A.L. Mackay, Gareth Thomas, J.W. Cahn, P. Ramachandra Rao, K. Chattopadhyay, D. Shechtman, K.H. Kuo and A. Inoue. Discussions with T.A. Abinandanan and Abhik Choudhury were stimulating. The work of Sudarshna Kalyanaraman on HEA is also acknowledged with appreciation. Grateful thanks are expressed to members of his family for support in writing this book.

The authors are deeply indebted to the meticulous reading of the entire book by Dr. R. Krishnan and thank him profusely for his useful suggestions.

B.S. Murty, J.W. Yeh and S. Ranganathan

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CONTENTS

CHAPTER 1

A Brief History of Alloys and the Birth of High-Entropy Alloys

1.1 INTRODUCTION

Alloying is the greatest gift of metallurgy to humankind. The English language insists on unalloyed pleasures, thereby implying that the sensation of pleasure must be pure and not admixed with other emotions. Exactly the opposite rules in metallurgy, where pure metals have few uses but lot more upon alloying. The power of this idea of alloying is not confined to metals. The same principle of alloying applies in polymers and ceramics. It can be carried further by mixing two classes of materials to create a variety of composites.

The civilizational journey of humankind began with the discovery of native metals such as gold and copper as pure metals. Nowadays we have access to an incredible number and variety of materials. Ashby map (Ashby, 2011) shown in Figure 1.1 gives a panoramic view of the development in the use of materials over 10 millennia. A graphic depiction of the different classes of materials from ceramics to metals, polymers, and more recently to composites is vividly displayed. The passage from discovery through development to design of materials can be noted. Ashby's (2011) map in term of strength versus density shown in Figure 1.2 demonstrates the filling of material–property space in a vivid fashion from 50,000 BCE up to the present scenario. In time scale, the largest filling has occurred in the past 50 years during which envelopes of metals, ceramics, and composites had a large expansion, and new envelopes of synthetic polymers and foam materials take a significant space. But, the filled area also seems to approach some fundamental limits beyond which it is difficult to go further (Ashby, 2011).

In many ways, the history of alloying is the history of metallurgy and materials science. Books and treatises have been written. An elegant and brief history is by Ashby (2008). Cahn (2001) has offered a magisterial survey of "The coming of materials science". Ranganathan (2003) wrote on alloyed pleasures—an ode to alloying. In the following sections, a few episodes in this epic journey are covered.

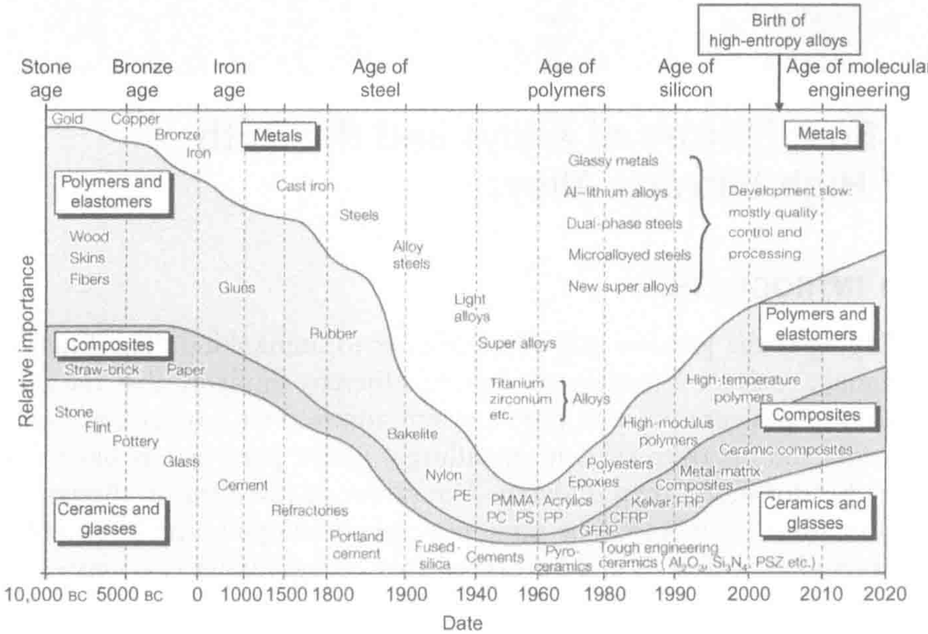

Figure 1.1 Historical evolution of engineering materials—marked with the birth of HEAs published in Advanced Engineering Materials *(Yeh et al., 2004b).* Adapted from Ashby (2011).

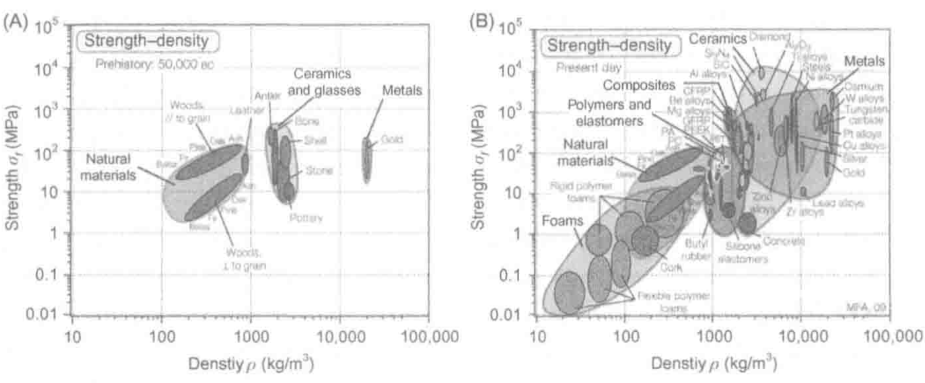

Figure 1.2 The explosion in the diversity of materials in the modern era (Ashby, 2011); (A) prehistoric era (50,000 BCE) and (B) current status.

1.2 THE COMING OF ALLOYS

Native alloys such as tumbaga and electrum are alloys of gold–copper and gold–silver, respectively. When platinum was discovered in 1735, it was compared with silver. Also, mixtures of platinum metals are found to occur in nature. It is an early example of multicomponent

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