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Light-Weight and Flexible High-Entropy Alloys

Yasong Li and Yong Zhang

Abstract

The lightweight and flexible materials can improve people's quality of daily life; in addition, the materials can be widely used in aerospace, automotive, consumer electronics, etc. Recently, high-entropy alloys had become hot issues in materials science with many excellent properties; therefore, we can combine the design ideas of high-entropy alloys with lightweight materials and flexible materials, taking into account the advantages of two types of materials, and promoting the development and progress of new materials. In the chapter, we will elaborate on the relationship between the microstructure and properties of lightweight high-entropy alloys and the design ideas of high-entropy alloys with flexible materials that were investigated in recent years. Furthermore, as the microstructure and mechanical properties of the alloys exhibit the nonlinear behaviors with entropy on high-entropy alloys, we would like to define the lightweight high-entropy alloy as the density is lower than 6 g/cm^3 , the mix-entropy of these alloys is higher than $1R$ (here, R is gas constant), and the number of components is four or more. Finally, it is expected to broaden the research field of high-entropy alloys and provide some new directions for the development of new materials.

Keywords: lightweight, high-entropy alloys, solid solution, alloy design, flexible materials

1. Introduction

Materials have always been a necessary progressive factor in human development; the progress of human society is often accompanied by advances in materials. From the Stone Age to the Bronze Age and then to the Iron Age, the emergence of each new materials has brought major changes in people's productivity. Nowadays, a series of materials have been used in various fields. The traditional structural materials such as steel, aluminum alloys, titanium alloys, magnesium alloys, etc. were still the most widely used materials. However, these materials cannot be applied in some specific areas. In addition, new materials have been developed such as composite material, nanostructure materials, carbon materials, bulk metallic glasses, high-entropy alloys, etc., as high-entropy alloys have been developed since 2004 by Yeh et al. [1] and Cantor et al. [2]. Due to the extremely complex composition of these alloys, the alloys also exhibit excellent properties that are difficult to achieve with many conventional alloys, such as high strength, high hardness, high fracture toughness, corrosion resistance, high temperature oxidation resistance, good low temperature performance, etc. In recent years, these high-entropy alloys such as AlCoCrFeNiCu, CoCrFeMnNi, CoCrFeNi (Ti, Al), NbMoTaW, CoCrNi (AlSi), etc. have been developed and studied [3, 4]. As these alloys also have large proportion of

transition metal elements, they also show high density. However, lightweight materials in the aerospace, automotive (especially electric vehicles), consumer electronics, and other fields have become an important development direction. However, designing novel lightweight materials with the concept of high-entropy alloys has become a hot issue, which will promote the development and applications [5, 6].

In view of the excellent performance of high-entropy alloys, we firmly believe that the lightweight high-entropy alloys have superior performance than traditional lightweight materials such as aluminum alloys, titanium alloys, magnesium alloys, etc. The general definition of lightweight materials generally uses the density of titanium alloy as the limit. The existing elements with lower density than titanium (4.51 g/cm^3) are mainly lithium (0.53 g/cm^3), beryllium (1.85 g/cm^3), boron (2.46 g/cm^3), sodium (0.97 g/cm^3), carbon (2.26 g/cm^3), magnesium (1.74 g/cm^3), aluminum (2.70 g/cm^3), silicon (2.33 g/cm^3), potassium (0.86 g/cm^3), calcium (1.55 g/cm^3), yttrium (2.99 g/cm^3), rubidium (1.53 g/cm^3), strontium (2.64 g/cm^3), strontium (4.47 g/cm^3), barium (3.51 g/cm^3), etc., and most of these elements are main group elements, which tend to have a higher chemical activity, with larger atomic radius, also with large difference in melting point and boiling point (lower melt point such as rubidium 39.3°C and higher melt point as titanium 1668°C). Also as we design the lightweight high-entropy alloys, these elements are not exactly used for the new alloy systems. Therefore, the development of lightweight high-entropy alloys often shows more difficulty than that of traditional high-entropy alloys.

In addition, compared with rigid materials, flexible materials are also widely used, which include foils, fibers, films, ribbons, etc., and usually they are made of organic matter. The inorganic materials such as silica, bulk metallic glasses, and metal materials, etc. tend to exhibit the characteristics of rigid materials. However, after being made into fibers or films, such materials can often undergo bending deformation due to the size effect and can also exhibit the characteristics of flexible materials. Nowadays, there is an increasing demand for flexible electronic materials in the field of electronics, especially in the field of wearable electronics. High-entropy alloys have demonstrated excellent overall performance as a new class of alloy materials in the field of rigid materials. Combined with the design concept of high-entropy alloy, can high-entropy open up a new research field in terms of flexible materials?

Nowadays, some scholars have also carried out a lot of research works; therefore, we will give a brief review on the relevant research works (mainly based on the research works of our own research group) and put forward our own opinions on the design and preparation of lightweight high-entropy alloys and high-entropy flexible materials.

2. Lightweight high-entropy alloy systems

2.1 Al-Mg-Li lightweight high-entropy alloy systems

Nowadays, the most commonly used lightweight metal materials are aluminum alloys, titanium alloys, magnesium alloys, etc. As the lithium alloys is the lightest structural metal material, which magnesium and aluminum are the common lightweight metal materials, our group firstly design the two lightweight high-entropy alloys systems (AlLiMgZnCu and AlLiMgZnSn) by Yang et al. [7].

With the design concept of traditional high-entropy alloys, we hope to form a multicomponent solid solution by alloy design. In recent years, these factors such as ΔS_{mix} , ΔH_{mix} , δ , Ω , $\Delta\chi$, VEC, T_m , etc., have made significant effects on the formation of solid solution as the design of high-entropy alloys. Therefore, in order to

Alloy design elements	Al	Li	Mg	Zn	Cu	Sn
r (10 ⁻¹⁰ m)	1.43	1.56	1.60	1.39	1.28	1.55
A (g/mol)	26.98	6.94	24.31	65.39	63.55	118.7
Crystal structure	FCC	BCC	HCP	HCP	FCC	Tetragonal
χ	1.61	0.98	1.31	1.65	1.90	1.96
VEC	2	1	2	12	11	4
ρ (g/cm ³)	2.70	0.54	1.74	7.13	8.93	7.37
T _m (K)	933.5	453.7	922	692.7	1358	505.1

Table 1.
Atomic radius (r), standard atomic weight (A), crystal structure, electronegativity (χ), value electron concentration (VEC), density (ρ), and melting temperature (T_m) for constituent elements in present alloys [7].

enhance the formation of the solid solution in lightweight high-entropy alloys, we firstly considered these factors and made some relevant calculations, which are shown in **Table 1**.

We had designed six lightweight high-entropy alloys that were AlLiMgZnSn, AlLi_{0.5}MgZn_{0.5}Sn_{0.2}, AlLi_{0.5}MgZn_{0.5}Cu_{0.2}, AlLi_{0.5}MgCu_{0.5}Sn_{0.2}, Al₈₀Li₅Mg₅Zn₅Sn₅, and Al₈₀Li₅Mg₅Zn₅Cu₅, and the densities of these alloys are 4.23, 3.22, 3.73, 3.69, 3.05, and 3.08 g/cm³, respectively; the density of these alloys is lower than that of titanium. The XRD pattern analysis of these alloys shows that the single-phase solid solution did not appear as the main phase under the condition of high mixing entropy; however, a large number of intermetallic compounds are produced during the smelting process. Only when the addition of aluminum reach to 80 at.%, the alloys show in a single face center cube(FCC) solid solution as the aluminum alloys. The SEM photos of these alloys which are shown in **Figure 1**, we can see a lot of intermetallic become the main phase of these alloys with high entropy, these show that the entropy did not victory when competition with the enthalpy, the solid solution did not form, also a lot of crack were found in the compounds, which cause the plasticity of these alloys are poor, also we when the aluminum become the main element of these alloy the α -Al (FCC) solid solution become the main phase in dendrite, some compounds which were rich in Cu or Sn in inter dendrite. The compression test of the alloys is shown in **Figure 2**, and the Al₈₀Li₅Mg₅Zn₅Sn₅ and Al₈₀Li₅Mg₅Zn₅Cu₅ alloys show good strength with higher than 800 MPa and yield strength higher than 400 MPa, with compressive plasticity better than 15%.

Also, the rare-earth elements lanthanum and cerium were added in these alloys to improve the solid solution formation ability of the alloys. In addition, Bridgeman directional solidification technology is also used in these alloys. However, these do not work in these systems. In order to further understand the formation law of solid solution of these lightweight high-entropy alloys, the δ , Ω , $\Delta\chi$, and VEC of these low-density high-entropy alloys are shown in **Figure 3**. Comparing the formation regions of solid solutions and intermetallic compounds with the conventional high-entropy alloys, we find that for the lightweight high-entropy alloys, which tend to have higher mixing enthalpy and electronegativity with smaller Ω and VEC, the δ of the alloy is in an intermediate region, often close to the critical region where the solid solution phase forms. In addition, the δ - $\Delta\chi$ can be a better way to predict the phase formation ability of these alloys. When $\Delta\chi < 0.175$, the solid solution will become the main phase of these alloys. Mainly, we found that the Al-Mg-Li system low-density high-entropy alloys had high chemical activity, which made it easier to form intermetallic compounds with other elements. Finally, we found that with the study of composition design, microstructure performance, and phase

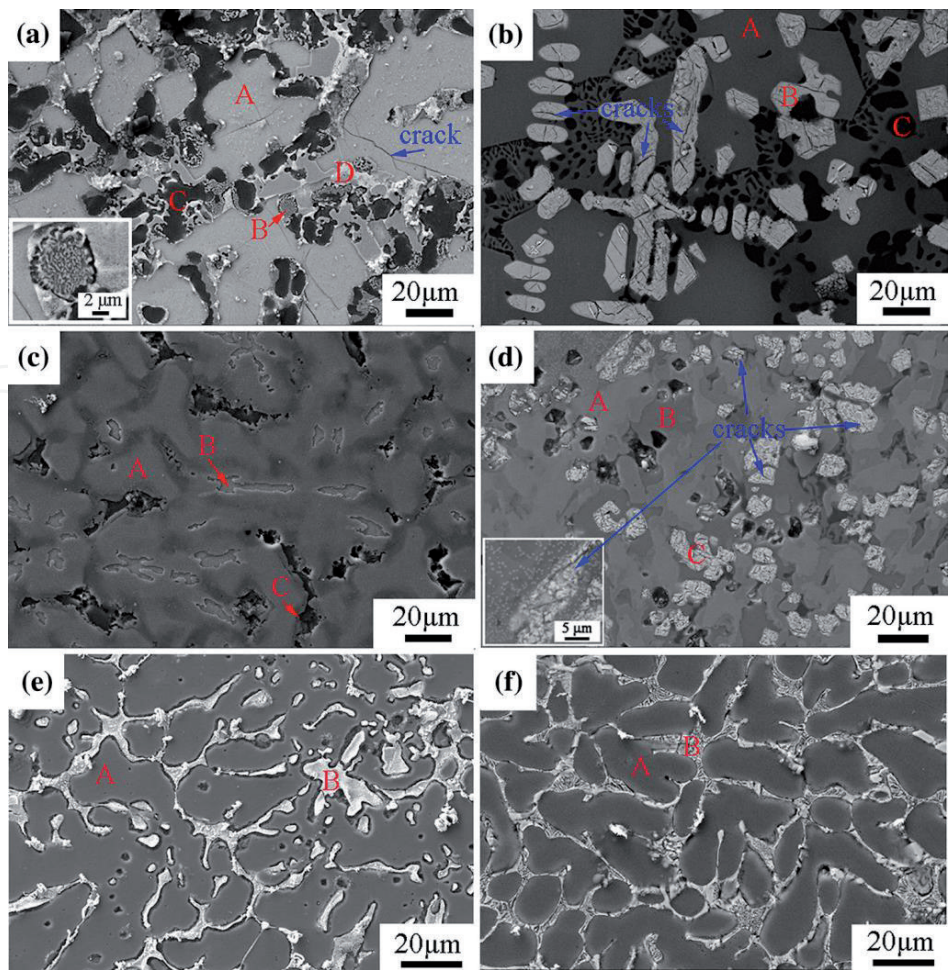


Figure 1. SEM secondary electron images of low-density multicomponent alloys. (a) AlLiMgZnSn; (b) AlLi_{0.5}MgZn_{0.5}Sn_{0.2}; (c) AlLi_{0.5}MgZn_{0.5}Cu_{0.2}; (d) AlLi_{0.5}MgCu_{0.5}Sn_{0.2}; (e) Al₈₀Li₅Mg₅Zn₅Sn₅; and (f) Al₈₀Li₅Mg₅Zn₅Cu₅ alloys [7].

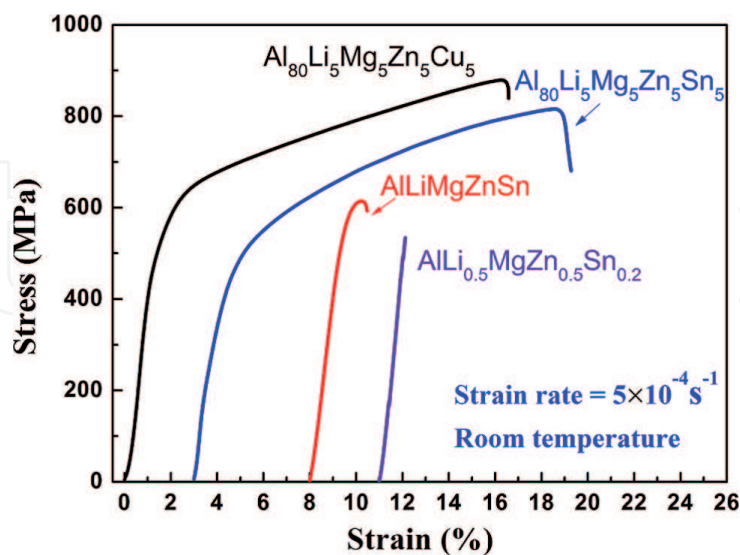


Figure 2. Compressive engineering stress-strain curves of AlLiMgZnSn, AlLi_{0.5}MgZn_{0.5}Sn_{0.2}, Al₈₀Li₅Mg₅Zn₅Sn₅, and Al₈₀Li₅Mg₅Zn₅Cu₅ alloys at room temperature. The initial strain rate was $5 \times 10^{-4} \text{ s}^{-1}$ [7].

formation of multicomponent alloys, for low-density high-entropy alloys, high mixing entropy is not the key factor in the formation of solid solution structures of these alloys. Compared with the traditional high-entropy alloys (mostly composed of transition metal elements), the solid solution phase formation conditions of the

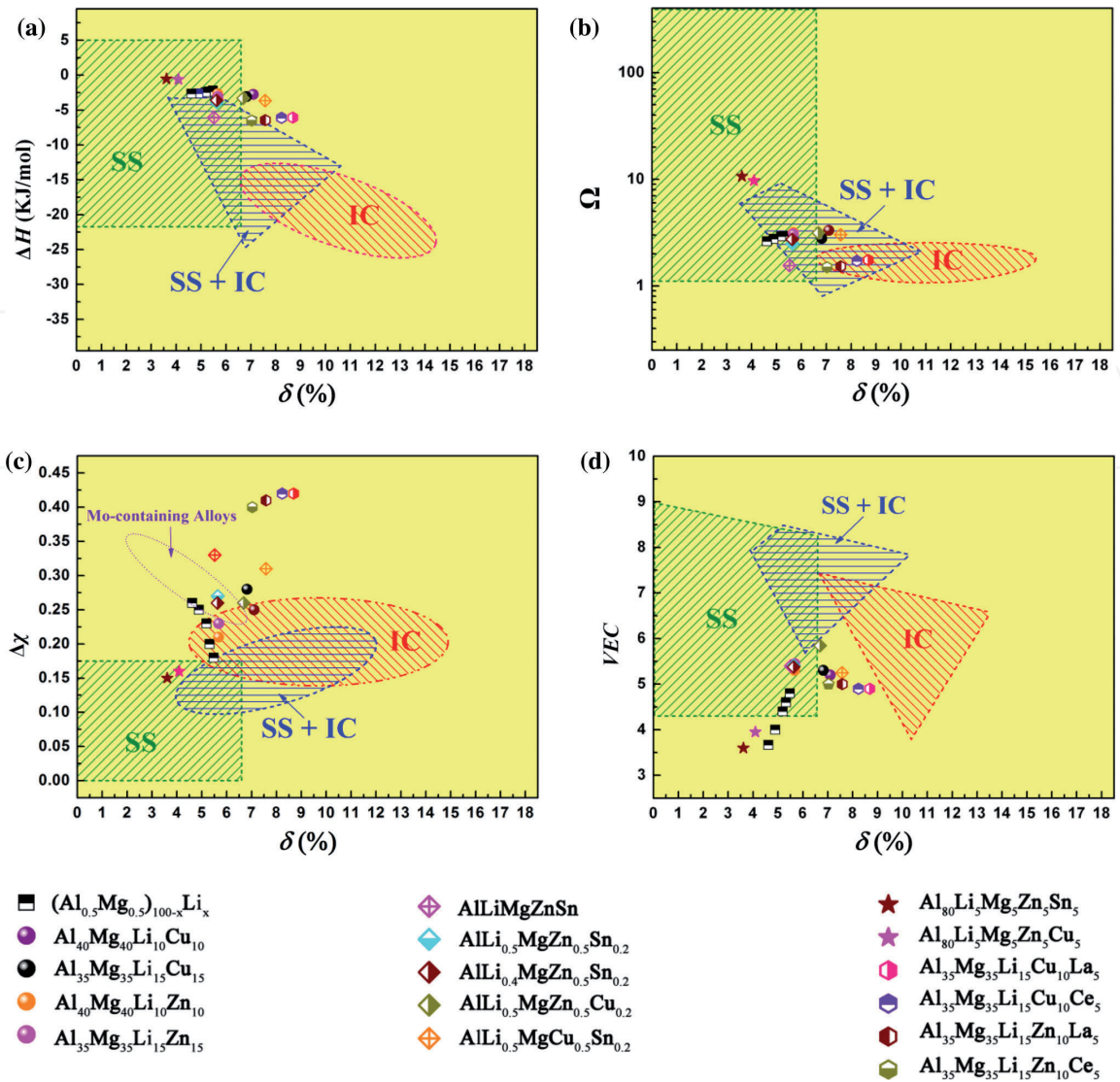


Figure 3.
Phase constituent prediction maps: (a) δ -Hmix, (b) δ - Ω , (c) δ - $\Delta\chi$, and (d) δ -VEC plots for multicomponent alloys in this work overlaid on cross-hatched regions developed in previous HEA investigations. (For $(\text{Al}_{0.5}\text{Mg}_{0.5})_{100-x}\text{Li}_x$, $x = 5, 10, 15, 25$, and 33.33) [7].

Al-Mg-Li-based lightweight high-entropy alloys are more severe. The solid solution formation of these alloys can be predicted by electronegativity ($\Delta\chi$); when the $\Delta\chi < 0.175$, it is easier to form the solid solution; and as $\Delta\chi \geq 0.175$, it tends to form the intermetallic compounds.

Li et al. also studied the microstructure and properties of the Al-Mg-Li high-entropy alloy system by using super-gravity technology [8]. Under different conditions with the super-gravity experiments, which found that supergravity does not separate the heavy elements of the alloy from the light elements; however, the microstructure of the alloy changed, which caused different properties. The alloy structure is still composed of α -Al solid solution structure and intermetallic compounds, and with supergravity, the microstructure changes to the eutectic microstructure. As supergravity is one entropic force, there are a variety of entropic forces in the process of alloy during solidification. Since gravity increases with distance, there are pressure and viscosity gradients in the molten metal. Meanwhile, due to the high mixing entropy of the alloy and the combination of various factors, the microstructure of intermetallic compounds and solid solution will change during solidification. In addition, these effects also made the grain refinement of the alloy along the direction of gravity to a certain extent, resulting in an enhancement of the strength of the alloy. Nevertheless, the alloy still does not form a single-phase

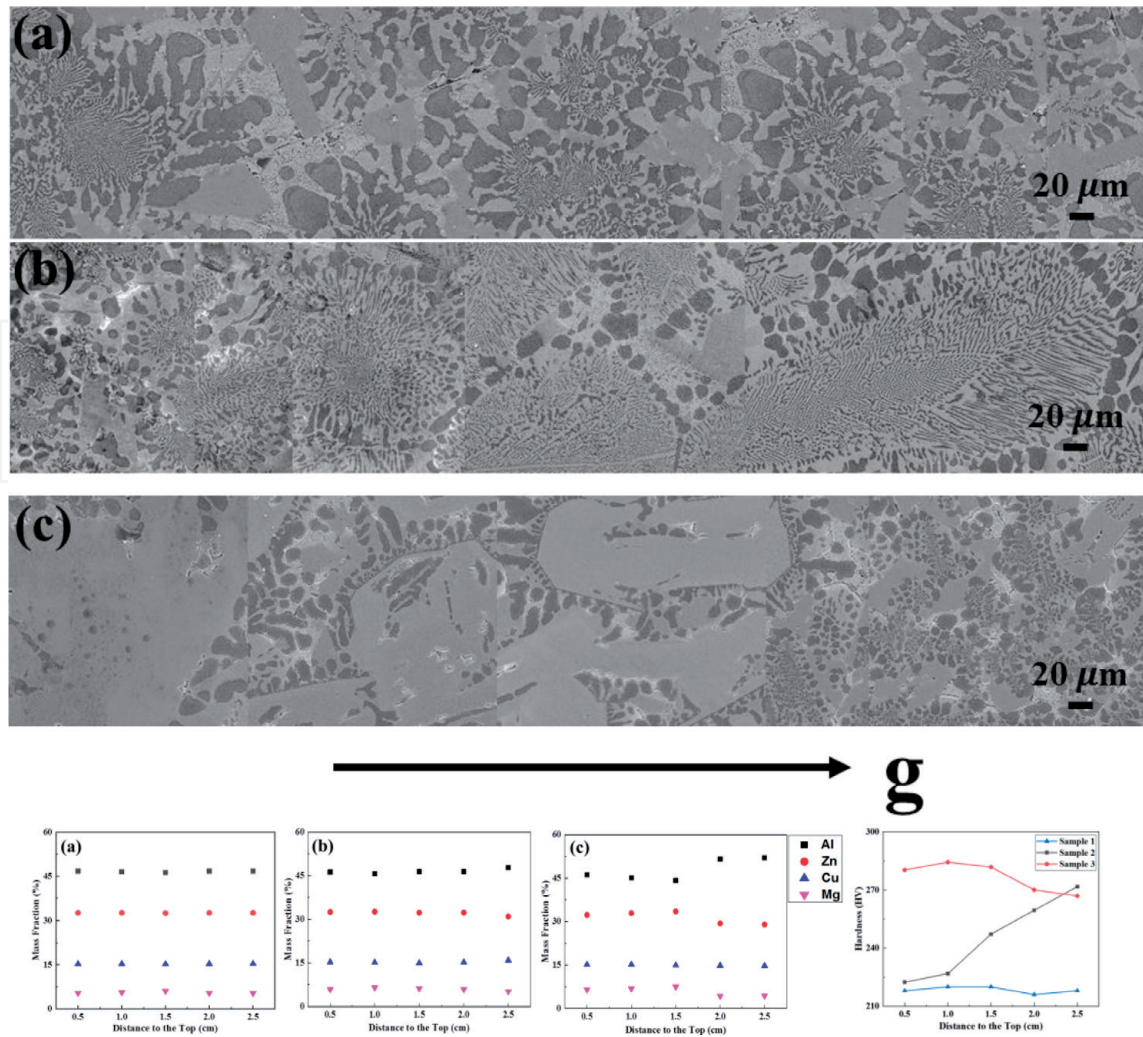


Figure 4.

Microstructure of the content of different element and hardness of the $\text{AlZn}_{0.4}\text{Li}_{0.2}\text{Mg}_{0.2}\text{Cu}_{0.2}$ alloy with different supergravity experiments: (a) Sample 1; (b) Sample 2, and (c) Sample 3 [8].

solid solution structure; therefore, the optimal structure of the alloy is the eutectic structure with the intermetallic compound and the solid solution with grain refine. **Figure 4** shows the microstructure, the composition of different elements by X-ray photoelectron spectroscopy, and the hardness of the alloy with the distance of gravity.

2.2 The Al-Mg-Zn-Cu-Si lightweight high-entropy alloy system

Based on the Al-Mg-Li study, our research group Shao et al. [9] used the Si exchange of Li, in order to reduce the cost of the alloy and expected to achieve lightweight, low-cost, high-entropy alloys; therefore, we studied the Al-Mg-Si system lightweight high-entropy alloys. Based on $\Delta\chi$, we designed the AlMgZnCuSi alloys, and these alloy samples were prepared by vacuum induction melting. We have found that the alloy forms a eutectic structure of solid solution and intermetallic compound when the content of Al is less than 80 at.%; however, these alloys show high strength with low ductility, and as the Al condition is higher than 80 at.%, they become α -Al face center cube solid solution. These alloys also have high strength with good compressive ductility. Which found that the $\text{Al}_{85}\text{Mg}_{10.5}\text{Zn}_{2.025}\text{Cu}_{2.025}\text{Si}_{0.45}$ alloy shows good toughness when the strength is higher than 800 MPa with ductility more than 20%. Currently, $\Delta\chi$ also predicts the phase formation of the alloy. We also found some serrated flow phenomena in the compressive strain curve of

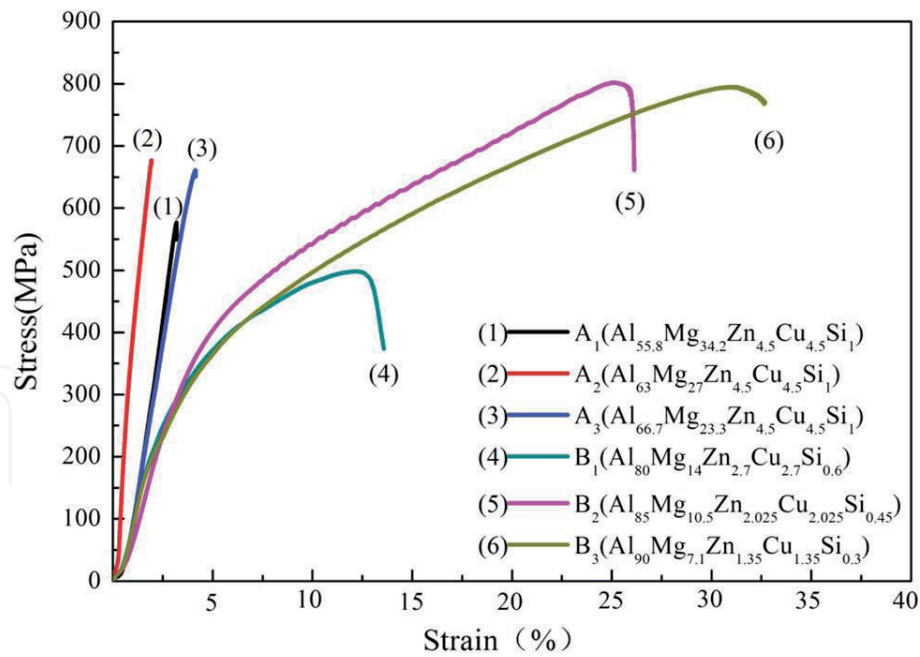


Figure 5.
 The compressive stress-strain curves at room temperature [9].

the alloy and will do some further research on the mechanism of serration behavior with the alloy. This research shows that this inexpensive alloy system is the research direction of another high-strength lightweight high-entropy alloy. The compressive stress-strain curves of a series of alloys at room temperature with a strain rate of 10^{-3} s^{-1} are shown in **Figure 5**.

In addition, some other researchers have also studied similar alloy systems based on this type of lightweight high-entropy alloy system. Baek et al. [10] used the ultrasonic melt treatment to prepare lightweight $\text{Al}_{70}\text{Mg}_{10}\text{Si}_{10}\text{Cu}_5\text{Zn}_5$ alloy, this alloy also forms a large number of other precipitated phases in the aluminum matrix, and the effect of solution treatment of this alloy on the microstructure and properties of the alloy was investigated, which found that the alloy has an excellent performance at both room temperature and 350°C ; however, the microstructure of the alloy is finely refined by the precipitation phase size by ultrasonic melt treatment technology, and mainly due to the introduction of trace amounts of Ti, the grain size is refined. In addition, mechanical properties of the alloy at room temperature have been improved with the solution treatment at 440°C , but the mechanical properties with high temperature (350°C) deteriorate. Through solution treatment, the Zn atoms redissolve into the second phases, which not only leads to the formation of fine super-saturated clusters in the matrix, but also spheroidizes the primary Si and Mg_2Si phases, thereby improving the room temperature mechanical properties of the alloy. They also studied the effects of $\text{Al-6Mg-9Si-10Cu-10Zn-3Ni}$ alloy aging treatment on properties and microstructure of alloys at different aging temperatures, and at 120°C , they found that the GP zone in the alloy with aging time was replaced by a Zn-rich metastable elliptical cluster to form a stable Zn precipitate containing a part of Cu atoms [11]. Besides, the aging precipitation behavior under different temperatures had also been studied [12]; as the aging temperature is below 70°C , a series of fine clusters and precipitates were formed, which greatly improves the strength of the composite. On the other hand, due to the coarsening of the precipitate, and the softening by the reduced volume fraction and the periodization of the second phase, a small strengthening effect was observed above 170°C . Sanchez et al. [13] have done some research on $\text{Al}_{65}\text{Cu}_5\text{Mg}_5\text{Si}_{15}\text{Zn}_5\text{X}_5$ and $\text{Al}_{70}\text{Cu}_5\text{Mg}_5\text{Si}_{10}\text{Zn}_5\text{X}_5$ systems and reported the effect of Fe, Ni, Cr, Mn, and Zr elements on the phase

formation, microstructure, and properties of these alloys. These researches all showed that this kind of alloy system has a good prospect in foundry industry.

2.3 High temperature application of lightweight high-entropy alloy

The light-weight metal elements as beryllium, scandium, titanium, yttrium etc., and the light-weight non-metallic elements such as carbon, boron, silicon etc., in addition to aluminum have a higher boiling point; therefore, these elements were also used for the design of high temperature application of lightweight high-entropy alloy. Some researchers have done a series of research work on these alloys.

Tseng et al. [14] studied the $\text{Al}_{20}\text{Be}_{20}\text{Fe}_{10}\text{Si}_{15}\text{Ti}_{35}$ lightweight high-entropy alloy with a vacuum-arc-melting, and this alloy showed a single hexagonal close-packed (HCP) structure solid solution phase, with high hardness ~ 8.9 GPa, high strength ~ 2.976 GPa, with a density of ~ 3.91 g/cm³; in addition, this alloy showed an excellent oxidation resistance at both 700 and 900°C, which is much better than the normal Ti-6Al-4 V alloy. Another way to prepare lightweight, high-temperature, high-entropy alloys is to reduce alloy density by adding lightweight elements Ti and Al to conventional alloys. These alloys usually have a higher density, however, lighter than the conventional superalloys, usually less than 6 g/cm³. These light-weight high entropy alloys, such as NbTiVTaAl_x [15], CrNbTiVZr [16], AlNbTiV [17], Al_{1.5}CrFeMnTi [18, 19], AlTiVCr [20, 21] etc., which tend to have a single-phase solid solution structure with lower density, good plasticity, and high temperature properties. In addition, these are a powerful alternative to the next generation of superalloys, with great potential to replace existing superalloys. The application of such alloys will bring a big leap in materials for the aviation industry.

2.4 Other lightweight high-entropy alloy systems

Finally, we will briefly explain the existing research on other lightweight high-entropy alloys. Youssef et al. [22] made an investigation on $\text{Al}_{20}\text{Li}_{20}\text{Mg}_{10}\text{Sc}_{20}\text{Ti}_{30}$ light-weight high-entropy alloy with mechanical alloying. Since such alloys were prepared by mechanical alloying, the alloy structure exhibited an ultrafine grain structure with only 12 nm, and the alloy exhibited an ultra-high hardness of 5.9 GPa, and its density was only 2.67 g/cm³; which shows a single face center cube (FCC) solid solution structure, when the power was milled without N, O, the alloy has a face centered cube (FCC) transformation into a hexagonal close-packed (HCP) structure with 500°C annealing treatment, however with N, O this transformation did not happen. Li et al. [23, 24] made an investigation on $\text{Mg}_x(\text{MnAlZnCu})_{100-x}$ lightweight high-entropy alloys, the microstructure of $\text{Mg}_{20}(\text{MnAlZnCu})$ alloy was consistent with HCP solid solution and Al-Mn icosahedral quasicrystal phase, and the compressive strength of these $\text{Mg}_x(\text{MnAlZnCu})_{100-x}$ alloys were high; however, the plasticity of alloys was poor. In addition, the microstructure and properties of the $\text{Mg}_{20}(\text{MnAlZnCu})$ alloy under different solidification conditions were also studied, and they found that with the faster cooling rate, the Al-Mn icosahedral quasicrystal phase was refined, which enhanced the strength of this alloy; however, with the brittleness of the HCP alloy, even the fast cooling rate can improve the plastic deformation ability of the alloy. The plasticity of this alloy is still poor, and with this work, they found that the high entropy can enhance the formation ability of icosahedral quasicrystal [24]. Du et al. [25] investigated the MgCaAlLiCu alloy, which shows a mainly single solid solution phase with tetragonal symmetry lattice structure, and the density of this alloy is ~ 2.2 g/cm³, with high compressive strength ~ 910 MPa. Jia et al. investigated the AlLiMgCaSi high-entropy alloys and they found that the density of these alloys were 1.46 to 1.70 g/cm³ and the strength was higher than 450 MPa, especially, as the $\text{Al}_{15}\text{Li}_{38}\text{Mg}_{45}\text{Ca}_{0.5}\text{Si}_{1.5}$ and $\text{Al}_{15}\text{Li}_{39}\text{Mg}_{45}\text{Ca}_0$

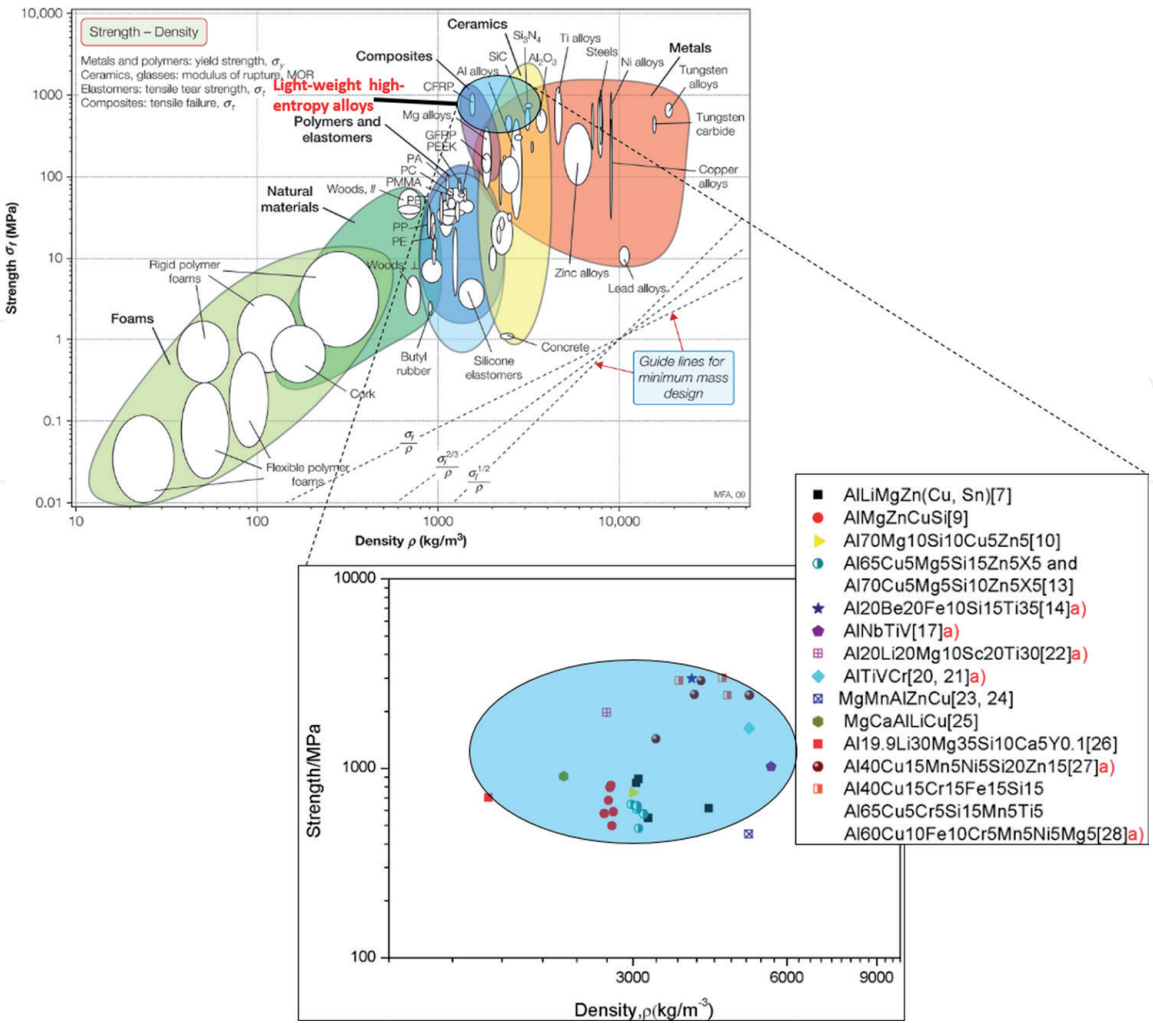


Figure 6.
The area of lightweight high-entropy alloys in the Ashby diagram of strength vs. density for structural materials [29], a) the strength of these alloys was equivalent as the hardness/3.

$_{5}\text{Si}_{0.5}$ alloy exhibited good plasticity ~45 and ~60%, which is much higher than most of the lightweight high-entropy alloys [26]. Sanchez et al. investigated on the as-cast high-entropy aluminums, and they found these alloys showed high hardness than other lightweight alloys [27, 28]. **Figure 6** shows the area of lightweight high-entropy alloys in the Ashby diagram of strength vs. density for structural materials, which we found that the strength of lightweight high-entropy alloys was much higher and the density much lower than some ceramics such as the SiC, Al₃N, etc.; however, the ductility is better than that of ceramics.

There are still many problems in the existing lightweight high-entropy alloys to be solved. First, the formation conditions of conventional high-entropy alloy solid solution need to be corrected. In addition, lightweight high-entropy alloys tend to exhibit high strength and poor room temperature plasticity, and we need to improve the toughness of these alloys.

3. High-entropy flexible materials

High-entropy alloys tend to have a solid solution structure, which means that these alloys had good plastic deformation capacity. The face center cubic (FCC) high-entropy alloys such as CoCrFeMnNi, Al_{0.3}CoCrFeNi, CoCrFeNi, etc. [30–36], show the excellent tensile plasticity which can exceed 50% at room temperature. Therefore, these alloys can be deformed by plastic deformation such as rolling,

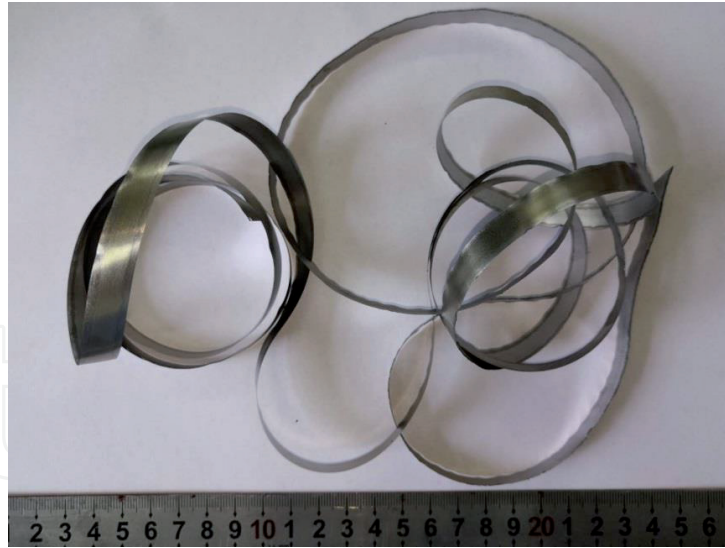


Figure 7.
High-entropy alloy ribbons by vacuum suspension quenching.

extrusion deformation, drawing deformation, etc., which can be made into foils, ribbon filaments, etc.; such materials tend to have the characteristics of flexible materials. In addition, further methods to obtain an alloy of a flexible material is the use of melt spinning method, or coating.

Ma et al. made the single-crystal structure the $\text{Al}_{0.3}\text{CoCrFeNi}$ alloy with the Bridgman solidification which found that the elongation of this alloy $\sim 80\%$, the alloy shows an excellent plastic deformation capacity [32]. Li et al. found that $\text{Al}_{0.3}\text{CoCrFeNi}$ alloy shows the elongation more than $\sim 60\%$ with forging [33]. Based on these studies the Li et al. formed the fibers with this alloy [34]. In addition, the high-entropy alloy ribbons and fibers can also be prepared by vacuum suspension quenching system, which Zhao et al. [35] use this technology prepared the $\text{CoFeNi}(\text{AlBSi})_x$ ribbons. High entropy alloy films can be prepared by chemical vapor deposition and physical vapor deposition, the thickness of these films tend to be $0.5 \mu\text{m}$ to $2 \mu\text{m}$, which become two-dimensional materials [37], also such films after separation from the substrate will be a flexible materials. Xing et al. [38] corrected the phase formation of the film materials with the concept of cooling rate. **Figure 7** shows the $\text{CoFeNi}(\text{AlBSi})_x$ high-entropy alloy ribbons by vacuum suspension quenching.

4. Conclusions

Summaries of the relevant properties and specificities of lightweight high-entropy alloys are as follows:

1. Lightweight high-entropy alloys are often limited by the addition of elements, and these elements tend to have high electronegativity. They are easy to form intermetallic compounds rather than solid solutions and it has been found that high mixing entropy does not promote the formation of solid solution phases.
2. Therefore, the concept of lightweight high-entropy alloy needs to be broadened, for which the mixing entropy $\Delta S_{mix} > 1R$ is a good choice.
3. Due to the excellent comprehensive mechanical properties of traditional high-entropy alloys, the lightweight high-entropy alloys are also supposed to have

great advantages. The density tends to locate between superalloys and titanium alloys, so we can broaden the density limit of lightweight superalloys, with a recommended density below 6 g/cm³.

4. Lightweight high-entropy alloys have broad application prospects. However, the development of lightweight high-entropy alloys has great problems. It is expected to make breakthroughs in this area by using some advanced design concepts and preparation methods.
5. Using the concept of high-entropy alloys, there may be new breakthroughs in the development of flexible materials.

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Conflict of interest

The authors declare no conflict of interest.

Author details

Yasong Li and Yong Zhang*
State Key Laboratory for Advanced Metals and Materials, University of Science and Technology Beijing, Beijing, China

*Address all correspondence to: drzhangy@ustb.edu.cn

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