High Entropy Alloys: Development and Applications

Steadyman Chikumba[1] and Veeredhi Vasudeva Rao[2]

Abstract**—**Traditional commercial alloys have been used for decades. The design of these alloys is based on selecting a main component otherwise known as a principal element depending on anticipated properties. The principal element would thus form the matrix. Commercially available alloy systems are either aluminium based, copper based, iron based or nickel based in the case of super alloys. However, this limits the number of alloys that can be made from these traditional alloy systems. New alloys have been developed at the turn of the new millennium referred to as High Entropy alloys. These alloys have been achieved through equiatomic substitution, by replacing individual components with multicomponent equiatomic or near-equiatomic mixtures of chemically similar species. Although vast information about the behaviour of binary systems is available, little knowledge is available about these multi-component systems. This paper reviews the development of high entropy alloys, their properties and characteristics and present and future applications.

Keywords**—**High Entropy Alloys, development, applications

I. Introduction

As various technological fields like aerospace, energy, transportation and manufacturing are advancing they present new demands on materials. New materials are being sought that can be able to meet new demands presented by operating conditions in these high end applications. High strength to weight ratio, creep strength and thermal resistance are among other desirable properties in these high end applications [1]. Traditional commercial alloys can achieve these functions but show limitations which include a lower specific weight and high density which impact negatively on structural applications [2] [3]. This paper reviews the development of High Entropy alloys their current and future commercial applications as substitutes to currently available commercial alloys.

II. Background

Metals account for about two thirds of all the elements and about 25% of the mass of the Earth [5]. Metals are rarely used in pure form, as they rarely exhibit a good combination of properties for thermal, structural and other mechanical applications [5]. This limitation in properties is overcome by alloying the pure metal with other elements [5]. Traditional alloys are thus based on adding solutes to a single base [6]. Currently there are thirty identifiable alloy systems with principal elements which include iron, magnesium, titanium, copper among others [1]. Alloys that have been developed contain one or two major elements selection of which is based on properties required by the alloy application [6]. Metallic alloys are thus able to exhibit a range of various properties and characteristics, such as strength, ductility, toughness, corrosion resistance, heat resistance, thermal expansion, thermal and electrical conductivity [1] [4]. [6]. Nickel based super alloys have been applied in the past 70 years, in high temperature load bearing applications since they are able to withstand temperatures of up to 1100^{o}C [2]. Other attributes they exhibit include good room temperature ductility, good creep and fatigue behaviour [2].

A. Composition, microstructure and properties

The influence of composition and microstructure of alloys on their properties and applications is a subject of research interest [8]. The composition of alloying elements influence an alloy's behaviour, properties phase constitution and structure or microstructure [6] [8]. Alloying elements have the capability to block slip planes hence improve mechanical strength [5]. In many applications of desirable alloy properties include high hardness, wear resistance, high temperature softening and oxidation resistance, anticorrosion résistance and high conductivity [7] [2]. However there are limitations to the mechanical, thermal stability at elevated temperature, density and wear properties of conventional alloys [8]. Properties of some alloys are summarised below in Table 1.

High strength and temperature commercial alloys are based on Nickel, Iron, Cobalt and Chromium, titanium among others [9] . Different alloys have their advantages and disadvantages. Titanium alloys are important for cryogenic applications while austenitic steel based on the Fe-Cr-Ni alloy system resist corrosion. Aluminium alloys have low density but low strength while Nickel alloys can offer high operating temperatures although they have high densities [2]. Some characteristics are shown below in table 1 [2].

Strength in metals and alloys depends on the distribution of a second phase eg β/ alpha titanium often in an intermetallic phase [2]. There are many applications where physical and mechanical properties are no easily met by traditional alloys [9].However, various methods can be used to improve alloy strength. These include solution strengthening, precipitation hardening martensitic hardening, carbide or added oxide strengthening and work hardening [9].

[1]Steadyman Chikumba is with Universirty of South Africa Mechanical and Industrial Engineering.

[2]Veeredhi Vasudeva Rao is with University of South Africa Mechanical and Industrial Engineering .

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TABLE I
PROPERTIES OF AL, TI AND NI ALLOYS

Alloy System	Density (gcm^{-3})	E (GPa)	σy (MPa)	Temp °C
Al	2.6-2.9	70	250-550	<150
Ti s	4.4-4.6	100-120	800-1400	<150
Ni	8-9	210-220	400-1300	<1100

Often alloys that have high strength and thermal stability at high temperature are required.

III. DEVELOPMENT OF HIGH-ENTROPY ALLOYS

The development of high entropy alloys is important in solving the shortcomings of convectional alloys in applications where operating conditions of loading and temperature are extreme [8]. The pioneering work on High Entropy alloys was in Taiwan, with the first findings on these alloys reported in 2002 [8, 10]. High Entropy alloys are composed of at least five major metal elements and with compositions that can give a range of properties [10] [11].Their definition was also extended to be an alloy with four components and one of the elements may exceed 35% [12]. Composition of minor elements may not exceed 5% [13].

Generally when a solid solution contains a high number of principal elements it has greater tendency to form solid solutions and not intermetallics or other phases [13] . This is the basis for the design of High-entropy alloys. Unlike the design of traditional alloys which is based on adding solutes to a single 'base' element, High Entropy alloys are designed by choosing elements that will form solid solutions when mixed at near-equiatomic concentrations [13]. Dependent upon their composition and processing route, High Entropy alloys possess a wide range of microstructures and properties [12]. Those containing Fe, Mn, Cu, Cr, Co and Ni are similar to austenitic stainless steels and perform well in structural applications [12].

A. Entropy of configuration

The formation of high entropy alloys can be analysed thermodynamically [10] [11].

According to Richard's rule molar entropy of fusion for most metals at melting temperatureT_m.

$$\Delta S_f = {\Delta H_f}/{T_m} = R \qquad (1)$$

At melting point entropy is high due to higher degree of randomness. Thus this lowers Gibbs free energy of the liquid or solid [8, 12]. From a thermodynamic point of view, when liquid and solid solutions are formed the free energy of mixing is given by

$$\Delta G_{mix} = \Delta H_{mix} - T\Delta S_{mix} \qquad (2)$$

$\Delta H_{mix}$is the enthalpy of mixing, $\Delta S_{mix}$is the change in entropy of mixing while T is the temperature.

For pure metals, at melting point entropy is theoretically equal to R the molar gas constant which is equal to 8.134 J/K mole, while for equimolar alloys it is higher than R as explained earlier [14]. The Boltzmann's hypothesis states that for a solid and liquid system containing n elements, the configurational entropy change per mole [8, 12]:

$$\Delta S_{conf} = -k\ln w = -R\left(\frac{1}{n}\ln\frac{1}{n} + \frac{1}{n}\ln\frac{1}{n} + \cdots\right)$$
$$= R\ln n \qquad (3)$$

where k is Boltzmann's constant, w is the number of ways of mixing, and R is the gas constant = 8.134 J/K mole [13]. ΔS_{conf} for equimolar alloys with 3, 5, 6, 9, and 13 elements are 1.1R, 1.1R, 1.61R, 1.61R, 1.79R, 2.2R and 2.57R respectively [11]. Therefore, the presence of more component elements in the alloy results in it having a higher the entropy of configuration. The relationship between change in configurational entropy and temperature can be used to classify alloys. This is shown in Figure 1 below. [12]

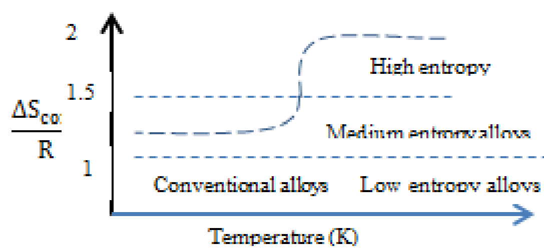

Fig. 1 Entropy versus temperature for alloys

Thus high entropy alloys and their atomic configuration are characterised by high mixing entropy phenomenon [12, 11, 15, 8]. Thus high entropy alloys are defined as alloys with a configurational entropy higher than 1.5R where R is the gas constant [13].

B. Selection of HEA constituent elements

Elements that can be used in High entropy alloys are shown below in Table 2 [11]. The main constituents of HEAs comprise metallic elements can be selected form metallic group and non-metallic elements which include C, B, Si, P, S H [11].

TABLE II
CANDIDATE MAJOR CONSTITUENT ELEMENTS FOR POSSIBLE USE IN HE ALLOYS

Major metallic elements	Minor metallic elements	Minor non-metallic elements
Li, Be, Mg, Al, Sc, Ti, V, Cr, Fe, Co, Ni, Cu, Zn, Y, Zr, Nb, Mo, Sm, Eu, Au, Gd, Tb, Rh, Pb, Pd, Ag, Hf Ta W Pt Nd	Li, Be, Mg, Al, Sc, Ti, V, Cr, Fe, Co, Ni, Cu, Zn, Ga, Ge, Sy, cd, Ln, Sn, Sb, Y, ZR< Nb, Mo, Ru, Rh, Pb, Bi, Pd, Ag, Hf, Ta, W, Pt, Au, La, Ce, Ce, Pr, Nd, Sm, Eu, Gd, Tb	C, B, Si, P, S, O, N

Jien-Wei et al [3] proved that an arbitrary choice of a group of 13 mutually miscible metallic elements enables the design of 7099 HE alloy systems with 5 to 13 elements in equimolar ratios, citing an example of CuCoNi-CrAlFe.

$$ {}^{13}_{5}C + {}^{13}_{5}C + {}^{13}_{6}C + {}^{13}_{7}C + {}^{13}_{8}C + {}^{13}_{9}C + {}^{13}_{10}C + {}^{13}_{11}C + {}^{13}_{12}C + {}^{13}_{13}C = 7099 \quad (4) $$

Thus, countless alloys can be produced even with elements that cannot produce chemical immiscibility [3]. Cantor [11] demonstrates the number of alloys that have been studied which include all unitary, most binary, and a few ternary and other alloy systems. In a system with c components, the number of independent components is $c - 1$.

If they differ by at least x% tolerance in at least one component each component can take on $(100/x)$ different possible compositions. The total number of different possible alloys N is:

$$ N = \left(\frac{100}{x}\right)^{c-1} \quad (6) $$

Elements in the alloying range of the periodic table, c = 40, and at a material specification, x = 1, gives a conservative estimate of number of possible alloys [11]:

$$ N = \left(\frac{100}{1}\right)^{39} = 10^{2x39} \approx 10^{78} \quad (7) $$

The number of alloys which have been investigated N_1 as considering sixty metallic elements in the periodic table of elements [11]:

$$ 60 < 60 + 60 + 59 \times \left(\frac{100}{0.1}\right) + 60 \times 59 \times 58 \times \left(\frac{100}{0.1}\right)^2 + \text{small} \approx 10^{11} \quad (8) $$

The proportion of all materials that have been investigated is [11]:

$$ \frac{N_i}{N} < \frac{10^{11}}{10^{78}} \approx 10^{-67} \quad (9) $$

Thus it can be concluded that there is a large number of exciting new materials yet to be discovered [11, 4]. In investigating how elements for alloying are selected as follows:

- Identify all metals for structural metals from literature and standards. Starting from the periodic table elements removing all non-metals.
- Identify desired attributes
- Use Hume – Rothery rules to find atoms within 8% of atomic size difference [4]. This important for cast alloys.

C. *Selection of a HEA alloy system*

Existing literature and standards can be used to identify candidate major constituent elements for possible use in the alloys. From elements in the periodic table with exclusion of radioactive ones possible candidates are shown in Table 2 [2] [11].

Candidate element selection is based on desired attributes elements such as temperature, density and strength among others. In terms of operating temperature material can be classified as Low-T (200°C), Medium-T (600°C) and High-T (1000°C) [2] [7]. Strength depends on microstructural properties while melting temperature depends on constituents, atomic sizes, and chemical interactions. The Pauling electronegativity and atomic radius is also important factor in selecting alloying elements [2]. Temperature of use must be 50% of temperature at which the alloy will be used [2]. The Hume – Rothery rules to find atoms within 8% of atomic size difference. This is important for cast alloys [2].

In single-phased HEAs it is difficult to reach a reasonable balance between strength and tensile ductility [16]. Research has shown that single-phased FCC structured HEAs are ductile but not strong enough [4]. They also exhibit inferior castability and compositional segregation, and this downgrades their mechanical properties and cast shadow on their engineering application [17]. Single-phased BCC structured HEAs can be very strong but at the price of brittleness [18].

In alloys systems with high mixing entropy there is a tendency of lowering ordering and segregation. This results in a more random solid solution being formed rather than intermetallics [3].

IV. METHODS OF MANUFACTURE OF HEAs

New processing technologies such as rapid solidification and mechanical alloying have aided the development in the manufacturing of multi principal element systems [1]. They can be manufactured using resistance melting, powder metallurgy, electric arc melting, rapid solidification, mechanical alloying, and induction melting [13] .

Arc melting happens in a sealed argon environment. The molten alloy is then cast. However, the challenge of heating the elements together like silver and copper is that the elements have a tendency to form a hypoeutectic which tends to separate itself from the rest of the elements [19].

Alloys can be made by powder metallurgy. An example is AlCo alloy which can be sintered [19].

When mechanical alloying is used it involves the milling of components elements together. This process results in amorphous and solid-solution phases as the two terminal phases [6]. The milling process can be first performed in a mixer cooled by liquid nitrogen which avoid welding of the powders. This is then followed by milling at room temperature [6].

HEAs can be also be made from chill casting or mechanical alloying. Cantor [11] investigated the structure of a wide range of chill cast alloys in the CrMnFeCoNiNbGeVTi system these alloys being based on the generic formula $Cr_xMn_xFe_xCo_xNi_xNb_xGe_xV_xTi_x$. Where there are high vapour pressures of several of the elements Li and Mg, mechanical alloying is used to prepare these alloys instead of melting and casting [6, 11].

Other techniques include arc melting, copper mould casting, Bridgman solidification. Optimisation of the microstructure is important and is achieved using rolling and heat treatments.

CrMnFeCoNi alloy can be prepared using high-purity elemental starting materials by arc melting and drop casting into copper molds [19]. This process is then followed by cold forging and cross rolling at room temperature into sheets. The resultant alloy after recrystallization, possesses an equiaxed grain structure [19].

V. PROPERTIES OF HEAs

HEAs show high resistance to softening at elevated temperatures and sluggish diffusion kinetics [9, 13]. The four core effects for HEAs are (a) Thermodynamics: high-entropy effects (b) Kinetics: sluggish diffusion; (c) Structures: severe lattice distortion; and (d) Properties: cocktail effects [13].

Severe lattice-distortion effect in HEAs is exhibited through severe lattice-distortion effect when compared with the one dominant element alloys, where the lattice site is occupied mainly by the dominant constituent. This is so because each element has the same possibility to occupy the lattice site, if ignoring chemical ordering. Since they are atoms of different sizes this leads to lattice distortion [19]. The cocktail effect implies that the alloy properties can be greatly adjusted by the composition change and alloying [20]. The change in properties with relative change in content is illustrated by the change in Al content in the $CoCrCuNiAl_x$ HEAs. With the increase of the Al con-tent, the phases change from FCC to BCC+FCC and then to BCC structures [12].

Eutectic HEAs are considered to be good candidate high-temperature alloys, since their near-equilibrium microstructures can resist change at temperatures as high as their reaction temperature, low-energy phase boundaries, controllable microstructures, high rupture strength, stable defect structures, good high-temperature creep resistance [21]. They form regular lamellar or rod-like eutectic organization, forming an in-situ composite [21].

HEAs of the AlCoCrFeNiTi family have high specific strength (ratio yield strength to density) compared to conventional alloys of titanium, nickel, steel and aluminium, natural polymers and bulk metallic glasses [12] [22] with a wide range of Young's modulus. They also exhibit good compressive strengths (400–500 MPa) at room temperature [6].

CrMnFeCoNi, which forms a single-phase face-centered cubic solid solution exhibits excellent damage tolerance with tensile strengths above 1 GPa and fracture toughness values exceeding 200 $MPa\cdot m^{1/2}$.It is also able to retain these properties at cryogenic temperatures as low as 77K [23].

An illustration of such superior properties in high strength high entropy alloys is shown by AlFeVSi high-temperature aluminium alloys in Table 3 [23]. The four component systems show high strength and thermal stability due to fine dispersion of incoherent intermetallic phases, oxides, and carbides [23].

TABLE III
HIGH TEMPERATURE ALUMINIUM ALLOYS [23]

Composition (wt%)	Density (g/cm^3)	Modulus (GPa)
Al-8.3Fe-4Ce	2.93	81.4
Al-7Fe-6Ce	2.96	78.9
Al-8.5Fe-1.3V-1.7Si	3.02	88.4
Al-12.4Fe-1.2V-2.0.3Si	3.07	95.5

Ingot Aluminium-Lithium alloys have even better structural efficiency, lower density and improved modulus of elasticity increasing structural weight by 7-15% and increasing elastic modulus by 10-20% [23].However there are limitations in their use for sheet metal applications which include anisotropy of mechanical properties, low ductility and fracture toughness [23]

VI. CURRENT AND FUTURE APPLICATIONS

Because of low density and high strength and find application in transportation and energy industries [2]. These applications require performance, reliability and endurance in extreme operating conditions [2]. Frictional heating can raise the surface temperature of supersonic aircraft to well above 300°C, and temperatures in the first- and second-stage compressor sections of turbine fan jet engines can exceed 400°C [23] . HEAs are good candidates to replace steel and Titanium alloys. Applications may include the compressor blades of an aero-engine which are often manufactured using Ti-base alloys.

HEAs can be used to protect the surface of machine components and tools because of their high hardness, wear resistance, high-temperature softening resistance, anti-corrosion, and combinations of these properties [8] [25] [26]. Hardfacing technology can be employed whereby the HE alloys are fabricated into rods and powders and then plasma arc or thermally-sprayed onto the surface of tools and other components [23]. The hardfacing process involves adding a thick layer of wear and or corrosion resistant material by welding, thermal spray welding. Common industrial uses include moulds, dies, tools and nozzles [24].

They are also increasingly finding applications as binders where particles such as Fe, Co and Ni, and ceramic particles such as WC and TiC [23]. AlCoX and CoCrX, have potential to replace the conventional binders in hard metals alloy binders after liquid -phase sintering, even without the presence of grain refiners as they have low contents of expensive cobalt and are composed of an FCC phase [24].

HEAs can be used in coatings used in food preservation and cook ware due to anticorrosion, anti-oxidation and wear resistance properties [23]. Bacteria for example E-coli can be prevented from reproducing and building colonies [23].

They can be used to suppress electromagnetic interference especially in electronics. For example at 13000MHz a coating of 1μm can be effective as a screen in commercial applications [23].

CrMnFeCoNi alloy can be used in cryogenic applications such as liquefied gas storage and can retain mechanical properties at temperatures as low as 77K. [19].

Thus there is wider scope for the application of high strength alloys.

VII. CONCLUSION

Although HEAs are still being widely explored, they have so for shown superior mechanical, thermal and chemical properties. These properties can lend them applications in structural, aerospace, food and energy industries. As the methods of manufacturing them evolve their costs of HEAs are going to come down which will allow them to find wider applications.

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