Prospect of high entropy alloys (HETAs) for advance application

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Abstract. Presently, HETAs are of enormous research significance in material engineering and sciences. In contrast to the traditional alloys with one and occasionally two base constituents, high entropy alloys consist of many essential elements, which are considered in excessive number compared to the conventional alloys. The arrival HETAs ignites some basic questions that challenge the hypothesis, mock-up, and preparation techniques of the traditional alloys. Here, the review of the latest studies on the prospect and essential subjects related to HETAs were provided. Besides, the novelty in the characteristics of high entropy alloys were also examined, which includes: high wear and corrosion resistance, outstanding mechanical behaviour at high temperature, superb service strength, and ductility, exceptional fracture toughness at extremely low or cryogenic temperatures, super-conductivity and super-paramagnetism. As a result of their richness of design, great structural and operational potential, they are now seen as promising materials for novel and advance applications, which should be further investigated.

Keywords: Alloys, Materials, Temperature, HETAs, Toughness

1. Introduction
From the primeval eras, the drive for conceptual design and development of new materials has been the goal of human society. Discovery of unknown metals and invention of novel alloys have played significant functions for more than hundreds of years. Ever since ancient times, the development of alloys has conventionally been by the base constituent or element concept [1, 2]. This alloying approach begins with one or sometimes two base constituents, such as the case of steels and superalloys with iron and cobalt, respectively, as the base elements, with some minor alloys for property enhancement. In recent years, there have been shifts in paradigm towards a proposed alloy design, which involves the mixture of several elements in an equimolar or near equimolar composition to produce alloys better than those of the based element approach [3]. These alloys of multiple elements are referred to as high-entropy alloys (HETAs), which imply that the alloys exhibit a high compositional degree of disorder due to the random incorporation of the elements. The single element principle of most conventional alloys gradually creates a severe embrittlement consequence with increasing strength, following the enduring dilemma of strength-ductility trade-off [4].

Currently, the studies on HETAs have been on the increase, even though the field is still being considered infant compared to the traditional alloying system [5]. Previously, in the design and development of traditional alloys, the focus of researchers was on a phase diagram’s corner, which is just a tiny portion of the design space. Conversely, the arrival of HETAs has prompted the shift in focus to the central section. Theoretically, this attribute is a drastic departure from the usual hypothesis and therefore provided fresh avenues for in-depth exploration of alloy design and development. HETAs were usually described as alloys with at least five key metallic constituents each contains an at.% ranging from 5 and 35 [6]. They can be single, double or multiple phases, each with a specific application. Single-phase HETAs exhibiting FCC structure originally attract enormous attention within the material community as a result of their numerous attractive mechanical characteristics such as outstanding tensile ductility and fracture toughness at extremely or cryogenic temperatures [7, 8].

Nevertheless, the low strengths at elevated and room temperatures make them unsatisfactory for many industrial applications. Hence, considerable attentions have been diverted towards dual and
multi-phase HETAs for accomplishing better strength and ductility combination. Although the matrix of single FCC HETAs alone is inadequately strong, their outstanding ductility and exceptional strain-hardening ability allow them to serve as a superb base alloy for further precipitation hardening [9, 10]. This strategy has been confirmed as one of the best approaches for strengthening metallic materials to be employed for structural applications both at elevated and room temperatures. In comparison with superalloys and steels, novel strengthened entropy alloys combine the advantages of ductile matrix and the micro-alloyed phase have shown great potential due to the superior combination of mechanical and physical characteristics, which has made them to increasingly attract research interest in recent years [11, 12]. In this review, the characteristics of HETAs were examined and the prospects for advance application were further highlighted.

1.1. Concept of HETAs
HETAs have been defined as alloys consisting of not less than five major elements. A good understanding of the likely interaction between mixing entropy and selection of phases is one of the subject matters of concern for researchers working on them [13]. Because of this, several questions have been asked on the mixing entropy in HETAs. Hence, the entropy of mixing per mole may be expressed configurationally as
\[ \Delta S_{\text{mix}} = - R g \sum_{i=1}^{E} M_i \ln M_i \]
where \( M_i \) is the molar fraction of \( i \)th element, \( R_g \) is the gas constant, and \( E \) is the total number of the elements composed in the mixture. It has been discovered from the contour diagrams of \( \Delta S_{\text{mix}} \) for multiple alloying systems that central region is usually the high entropy region where \( \Delta S_{\text{mix}} \) is much larger, whereas for the conventional alloys \( \Delta S_{\text{mix}} \) is smaller, and are indicated at the corner regions [14]. In most alloys of metal, the energy gain is adequate for the stabilization of entropy of a random solid solution phase against intermetallic complexes. Therefore, the entropy of mixing, \( \Delta S_{\text{mix}} \), for an equiatomic HETAs is given by
\[ \Delta S_{\text{mix}} = R g \ln E. \]
In accordance to the high entropy effect, the stabilization of the random solid solution against the inter-metallic complexes could be achieved when several constituents are mixed in an equimolar proportion [15, 16].

Since the HETAs normally appear to contain far lesser phases than Gibb’s phase law would permit, it has been suggested that compositional entropy provides major stabilization effect even in alloys of multiple phases. More so, many studies have not been able to substantiate phases present in HETAs, therefore, it has become necessary to carefully access the stability before the number of phases could be established. Although confirming an alloy’s absolute thermodynamic stability have been considered experimentally impossible, however, in the assessment of the stability of HETAs, there are important features that the studies should include: suitable heat treatments should be chosen to homogenize the as-cast alloy, and then promotion of phase decomposition. Heat treatment of as-cast HETAs produces near-stable microstructures and via cautious characterization of HETAs necessary assertion on their stabilities can be made [17]. Previous researches have revealed that most HETAs are made of face-centred cubic (FCC), body centred cubic (BCC), or combination of BCC and FCC solid solutions due to their high compositional mixing entropy. The configuration of these HETAs is ordered to achieve promising attributes such as high oxidation, corrosion and wear resistance as well as ductility and hardness [18, 19].

1.2. Techniques of production of HETAs
The processing techniques such as mechanical alloying and rapid solidification have promoted development in the production of multi-principal element structures. HETAs are produced using electric arc melting, mechanical alloying, induction melting, rapid solidification, powder metallurgy and resistance melting. Electric arc melting is commonly used; where the arc melting is performed a sealed argon atmosphere and followed by casting of the molten alloy. However, one main challenge of heating constituents like copper and silver together is their capacity to form a hypoeutectic, which attempts to detach itself from the other elements, and as such powder metallurgy, which involves sintering of alloys or element, can be employed [20].

In the production of HETAs from mechanical alloying, milling of constituent elements is involved. The product often contains two phases, referred to as terminal phases, which are the solid solution and amorphous phases. To prevent welding of powder, the milling process can be carried out first in mixer cooled using liquefied nitrogen. Some HETAs are made from cold casting, however, where the
vapour pressure of the numerous elements in the mixture is high, mechanical alloying is used for the preparation of the alloys instead of cast and melting. Other production methods are Bridgman solidification and copper mould casting [21].

2. Characteristics and prospects of HETAs

The properties of HETAs have been studied in severally, with all indication that the basic issues such as the thermodynamic root have not been totally resolved. However, they are reported to exhibit superior physical and mechanical characteristics as well as ultra-high fracture toughness greater than that of most alloys and pure metals, exceptional strength likened to some structural ceramics, excellent resistance to corrosion and high conductivity [22, 23]. These outstanding properties of HETAs have been broadly ascribed to [24, 25]:

- The effect of slow dispersion, which impedes the development of the cores of the second phase from a single-phase solid solution and consequently promoting the creation of nano-precipitates.
- The severe deformation effect of the lattice, which offers surplus strength and as well aids the slow kinetics in HETAs.
- The mixture effect, which could improve the characteristics via alloying.
- The effects of high entropy, which provide the thermodynamics for making stable the single-phase solid solutions.

The above descriptions do not only clarify the multifaceted occurrences that await explanations but as well offered valuable procedures for investigating the structure and behavioural relationship in HETAs.

2.1. Outstanding fracture toughness and ductility of HETAs at cryogenic and room temperatures

In most conventional alloys, an increase in strength has resulted in a reduction in ductility, while reductions in strength have yielded an increase in ductility. However, a lot of metallurgical researchers have made efforts to develop alloys that exhibit both high ductility and strength through grain refinement and gradient microstructures development with nano-twins. Alloys are known to exhibit stacking fault (SF), whose energy is significant in the evolution of twinning. Generally, the higher the SF energy, the more difficult the evolution of twinning. It is interesting to state that some HETAs possess very low SF energy, even extremely lower than some steels, and alloys of aluminium and copper [26].

In reality, a low SF energy promotes the division of complete dislocation to partial and therefore restraining cross-slip and climb dislocation. With a low SF energy, improved yield strength can be achieved for alloys, could result in twinning incited plasticity, thus leading to high ductility in suitable environments. It was recently discovered that some HETAs with FCC structure of single-phase and sizeable SF energy exhibit no nano-twinning when deformed plastically at room temperature. Some single-phase HETAs with low SF energy also possess great ductility, unlike the known conventional alloys. Interestingly, nano-twinning may possibly not be the only fortification mechanism for alloys, solid solution hardening has also played a vital role in alloys strengthening [27].

HETAs with Single phase and FCC structure have attracted numerous attentions as cryogenic materials for structural application due to their superior mechanical characteristics which include high ductility, great fracture toughness and high strength. They are also found to exhibit outstanding damage resistance with high fracture toughness values and ultimate tensile strengths comparable or even exceeding those of the most highly rated cryogenic steels such as high nickel steels and austenitic stainless steels. The advent of mechanical nano-twinning has been regarded as the main reason for the improved mechanical characteristics at room and cryogenic temperature [28]. However, the comparatively low yield strengths Single-phase FCC HETAs makes them unsatisfactorily tough for several crucial applications. Interestingly, L1₂-strengthened or γ’ (gamma prime) strengthened HETAs can possess higher yield strengths greater than or equal to 1 GPa at room and cryogenic temperatures. In like manner to the single FCC HETAs enhancement in ductility and strength could be observed at about 77 K deformation temperature, exhibiting extremely high (ultimate tensile strength) UTS, high (yield strength) Ys and improved ductility by about 50 per cent. Such an
unprecedented property was attributed to the active development of SF with nano spacing. The excellent strain-hardening potential, high Ys and the exceptional tensile ductility of HETAs with L12-strengthened phases make them highly attractive materials for cryogenic applications [29].

2.2. Superior mechanical property at elevated temperatures and other exclusive novelty in HETAs

As initially stated, ‘the evolution of single-phase solid solutions is promoted by the enormous mixing entropy in HETAs at elevated temperature’. This consequently makes the development of structural alloys for high-temperature application effortless. The exceptional mechanical characteristics of these alloys are achieved at elevated temperature due to slow diffusion of the elements contained in the mixture. Most existing high temperature single-phase refractory HETAs with BCC structure exhibit high yield strength that can be likened to that of conventional superalloys such as Haynes 230 and Inconel 718. Inconel 718 has a similar strength to the refractory HETAs at low temperatures. However, Inconel 718 thermodynamically softens significantly at a temperature above 600 °C, while refractory HETAs are able to still sustain their strength even above the temperature of 1200 °C [30].

Besides, most of the latest research work on HETAs has revealed that, apart from the exceptionally high-temperature performance of the refractory HETAs, their density can also be drastically reduced by replacing the heavy constituents present in the composition with lighter elements. The likes of W and Ta can be replaced with lighter elements like Al, Cr and Ti to reduce density. While density is been affected by these alloying element, the yield strength at ambient and elevated temperature is improved [31]. One of the most significant mechanical properties expected of materials to be used for high-temperature applications such as gas and steam turbines component is the creep resistance. The γ strengthened HETAs have been seen to exhibit exceptional creep resistance due to their fairly low SF energy, high thermal stability of γ precipitate and great creep activation energy. They were also found to possess higher rapture life compared to numerous advanced nickel-based superalloys subjected the same test conditions [32].

Apart from the advance mechanical properties of HETAs, a number of them have been found to possess fascinating and unique serviceable characteristics as a result of their high compositional entropy, which restricts precipitation at a nanometer scale if at all any takes place. Some HETA such as AlNiCo(CuFe) has been reported to possess super-paramagnetic behaviour [33], while some others exhibit super-conductivity performance at critical temperatures [34]. This indicates that HETAs can be employed for electro-magnetic and super-conducting applications like particle accelerators and magnetic resonance imaging.

2.3. Exceptional strength and hardness of HETAs

Engineering alloys are expected to exhibit low density, high hardness and outstanding strength. These characteristics engender the use of some alloys for structural and advance applications such as in automotive, aircraft, ships and engineering construction where regulation of materials density is crucial for minimizing the demanded operational energy. In respect of this, researchers have reportedly developed lightweight refractory HETAs with high strength. HETAs such as Al10B12Be20Fe10Si15Ti35 is developed from the mixture of low-density refractory constituents were found to exhibit very high hardness and strength better than most conventional and superalloy. The strength and hardness were ascribed to the availability of disordered solid solutions of BCC in the alloys. More so, some HETAs exhibit significantly high compressive yield strength at room temperature [35].

Recently HETAs with ultra-high-strength, super low densities and HCP structures are being developed through mechanical alloying. These alloys were discovered to be significantly harder than most conventionally developed alloys. However, the specific strengths are much time higher than those of conventional low-density alloys such as Cu alloys, Ti alloys and Zn alloys, but comparable to silicon carbide ceramics [36].

2.4. Oxidation resistance tendencies of HETAs

The oxidation resistance of metallic materials has been significantly improved via the addition of alloying elements such as aluminium (Al) and chromium (Cr). Although the oxidation properties of L12-strengthened HETAs have been sparsely, however, the conduction of isothermal oxidation of L12-strengthened HETAs at elevated temperature and over substantial time range indicates that no oxide spallation was present. High concentrations of different solutes in L12-strengthened HETAs have been
discovered to result in the development of complex oxides. HETAs are known to exhibit three main oxidation structures, which are: the outer layer, the inner layer consisting of a complex mixture of several compounds and the layer beneath, which often consists of the oxide of aluminium (Al₂O₃) and oxide of chromium (Cr₂O₃), resisting internal oxidation especially in HETAs whose mixture or composition contains aluminium and chromium. It is an established fact that Al₂O₃ provides more effective surface protection at elevated temperature compared to Cr₂O₃ due to the lower oxygen permeability and more thermal stability. Above 950 °C, Cr₂O₃ usually loses its stability and may slowly change to the unstable CrO₃. Basically, the outstanding oxidation resistance of L₁₂-strengthened HETAs at elevated temperatures is drawn from the formation of Al₂O₃ layer, which protects the material from hot oxidation products and preserve the metals inherent properties for advance operational performance [37, 38].

3. Present and future applications of HETAs

The novel HETAs have found applications as structural materials in nuclear and power plant, automobiles, electronics, submarines and aerospace industries. They have been used several for high-temperature applications. For the turbine fan of aircrafts engines and compressor blades, HETAs are potential materials which can replace the likes of Titanium and steel alloys and for higher efficiency since these applications require high-level reliability and extremely fatigue performance in operation. In supersonic aircraft, the frictional heat can shot the surface temperature above the temperature that material can accommodate, but with HETAs the heat is suppressed or absorbed due to the high fatigue and creep resistance characteristics. They can also be used for the protection of machine components and tools surfaces due to their high wear and corrosion resistance, high hardness, high creep and fatigue resistance, and combination of these dynamic characteristics. HETAs can be fabricated on the surface of tools and several components in the form of powder and rod, and then thermally sprayed or plasma arc to provide hard facing technology. This technology process involves the addition of thick film of corrosion or wear resistance materials via welding or thermal spraying for protection of nozzles, tool, moulds and dies [39].

More so, HETAs have been used as reliable binders. They have the potential to completely replace the usual binders after going through the liquid sintering phase, even in the absence of grain refiner and composed of FCC phase. They can be used as fillers in micro-joining because the traditional brazing technique results in cracking during the joining process of ceramic and metal components as a result of the availability of several interfacial complexes and the inherent residual stress. These interfacial complexes may be surmounted by the optimal balancing of strength-ductility of HETAs and the solid solution phase effect of the alloys. These alloys have been coated on cookware to enhance their wear, oxidation and corrosion resistance performance. The cryogenic of some HETAs can be explored in liquefied gas storage applications since they can retain their mechanical at extremely temperature [40].

4. Conclusion

This review has revealed that HETAs consists of the blending together of several elements, in contrast to the conventional alloys with one or occasionally two principal elements. HETAs are of immense significance due to the high wear and corrosion resistance, outstanding mechanical behaviour at high temperature, superb service strength and ductility, exceptional fracture toughness at extremely low or cryogenic temperatures, excellent strain-hardening potential, super-conductivity and super-paramagnetism they exhibit. These exceptional characteristics were attributed to L₁₂ or γ¹ strengthening phase and active development of SF with nano spacing. Basically, the outstanding oxidation resistance of L₁₂-strengthened HETAs at elevated temperatures is drawn from the formation of Al₂O₃ layer, which protects the material from hot oxidation products and preserves the metals inherent properties for advance operational performance. These properties engender the use of HETAs in nuclear and power plant, automobiles, electronics, submarines and aerospace industries. They can also be used in other applications like liquefied gas storage, machine components and tools protection, filler, binders and coating for cookware because of their great anti-oxidation, anti-corrosion and wear resistance prowess.
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