Near-equiatomic high-entropy decagonal quasicrystal in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$

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ABSTRACT High-entropy alloys (HEAs) contain multiple principal alloying elements, but usually with simple crystal structures. Quasicrystals are structurally complex phases, but are generally dominated by only one element. However, near-equiatomic high-entropy quasicrystals have rarely been reported because they are difficult to prepare experimentally and predict theoretically. Therefore, the preparation and crystal structures of near-equiatomic high-entropy quasicrystals have drawn much interest. We report a quinary decagonal quasicrystal (DQC) with near-equiatomic alloying elements in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons, which is the first to our knowledge. Meanwhile, the structural features of the DQC are characterized in detail. The configurational entropy of both the alloy and DQC satisfies the entropy-based criterion for HEAs, suggesting a high-entropy DQC. Our findings provide a new strategy to develop high-entropy quasicrystals.

Keywords: high-entropy alloys, decagonal quasicrystal, high-entropy quasicrystals, melt-spun ribbons, high-angle annular dark-field scanning transmission electron microscopy

INTRODUCTION

The first icosahedral quasicrystal was discovered by Shechtman in quenched Al-Mn alloys in 1984 [1], and hundreds of quasicrystals have since been reported [2]. Quasicrystals were mainly discovered in binary and ternary Al-based alloys [2–7], usually with an Al content of 60 at% to 85 at% (at%, atomic percent) [8]. In addition, quasicrystals were found in other systems such as Mg-based and Ga-based alloys [9–12]. Most quasicrystals are dominated by only one element; quasicrystals with multiple principal alloying elements have rarely been dis-

covered.

High-entropy alloys (HEAs) were first independently reported in 2004 by Yeh *et al.* [13] and Cantor *et al.* [14]. Different from the traditional alloys, HEAs are not based on one or two major elements, but are composed of multiple principal alloying elements. They usually contain at least five principal elements in equal or near-equal atomic percentages [15]. The definition of HEAs has been extended from equiatomic alloys to alloys with sufficiently high configurational entropy (ΔS_{conf}) to overcome the effect of enthalpy [16]. Alloys with a ΔS_{conf} greater than $1.5R$ ($R=8.31 \text{ J K}^{-1} \text{ mol}^{-1}$, the gas constant) are usually defined as HEAs [17,18], and non-equiatomic HEAs have also been developed [16].

The phase structures of reported HEAs contain solid solutions, intermetallics, and amorphous phases [19,20]. Among them, the face-centred cubic (FCC) [14,21–25] and body-centred cubic (BCC) [26–29] solid-solution HEAs are by far the most extensively investigated. Furthermore, hexagonal close-packed (HCP) structures [30–35], ordered intermetallic phases [16,36], complicated compounds [37], and metallic glasses [38] have also been observed in HEAs.

High-entropy quasicrystals are highly desired because they combine structural and compositional complexity. Although Takeuchi *et al.* [39] found an icosahedral quasicrystal in the heat-treated melt-spun amorphous alloys of $\text{Ti}_{33.33}\text{Zr}_{33.33}\text{Hf}_{13.33}\text{Ni}_{20}$ and $\text{Zr}_{30}\text{Hf}_{30}\text{Ni}_{15}\text{Cu}_{10}\text{Ti}_{15}$, the ΔS_{conf} of these alloys are $1.42R$ and $1.37R$, respectively. These are somewhat lower than $1.5R$, the criterion for HEA definition according to Yeh *et al.* [17,18]; thus, these alloys can be classified as medium-entropy alloys (MEAs)

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[15] instead of HEAs. Takeuchi *et al.* [39] considered $\text{Pd}_{20}\text{Pt}_{20}\text{Ni}_{20}\text{U}_{20}\text{Si}_{20}$ as a candidate for a high-entropy quasicrystal. However, uranium is a radioactive element, which is hard to handle in a conventional experimental environment. They also mentioned that it is difficult to explore high-entropy quasicrystals from Al-based quasicrystal alloys because there are no elements that can effectively replace Al in the periodic table [39]. Therefore, preparing high-entropy quasicrystals is still a great challenge.

We report a high-entropy decagonal quasicrystal (DQC) in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons, which is the first to our knowledge. Alloys for searching high-entropy DQCs are designed by referring to similar structural blocks of DQCs, the difference in atomic radius (δ) and the enthalpy of mixing (ΔH_{mix}). In general, the formation of a single-phase solid solution corresponds to the region of $-15 \text{ kJ mol}^{-1} < \Delta H_{\text{mix}} < 5 \text{ kJ mol}^{-1}$ and $0 < \delta < 6.6$ [16]. A further increase in δ and decrease in the negative ΔH_{mix} value result in the appearance of different phases such as intermetallic and bulk metallic glasses (BMGs) [16,40]. We found a similarity between Ga-Mn [41] and Al-Cr-Fe-Si [42] DQCs by comparing the 2-nm decagons of DQCs in different alloys. In addition, $\delta = 7.67\%$ ($> 6.6\%$) and $\Delta H_{\text{mix}} = -25.6 \text{ kJ mol}^{-1}$ ($< -15 \text{ kJ mol}^{-1}$) in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$, suggesting it is easier to form intermetallic compounds than solid solutions [16]. This is consistent with the features of intermetallic compounds of quasicrystals. Therefore, we selected the quinary Al-Si-Mn-Fe-Ga system for study and found a near-equiatomic high-entropy DQC with a periodicity of 1.23 nm in the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons. The structural characteristics of the high-entropy DQC were investigated in detail using high-angle annular dark field scanning transmission electron microscopy (HAADF-STEM) images at an atomic resolution.

EXPERIMENTAL SECTION

Materials

Al (99.99 wt%), Si (99.3 wt%), Mn (99.9 wt%), Fe (99.9 wt%) and Ga (99.9 wt%) metals were obtained from Beijing Cuibolin Nonferrous Metal Technology Development Center Co., Ltd. Copper grid was purchased from Beijing Zhongxingbairui Technology Co., Ltd.

Preparation of quasicrystals

An Al-Si-Mn-Fe-Ga alloy ingot of around 1 kg with a nominal composition of $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ was ob-

tained by first melting high-purity elements in an induction furnace under an argon atmosphere, and then pouring the molten alloy into a graphite crucible in the furnace to form an ingot. Several small sample pieces from the cast ingot of the alloy were subjected to the single-wheel melt-spun treatment under a copper wheel speed of 25 m s^{-1} , and then the melt-spun ribbons were obtained.

Characterizations

The specimens for TEM observation were prepared *via* three methods: powders spread onto a 3-mm-diameter copper grid covered with a holey carbon film, ion milling, and a focused ion beam (FIB). The secondary electron images were obtained with a ZEISS GeminiSEM 300 scanning electron microscope (SEM). The selected-area electron diffraction (SAED) patterns and energy dispersive X-ray spectroscopy (EDS) measurements were performed with an FEI Tecnai F30 transmission electron microscope. Meanwhile, the atomic-resolution structural investigations were performed on a Thermo Fisher Titan Themis microscope fitted with aberration correctors for the imaging lens and the probe forming lens, Super-X EDX system, operated at 300 kV. The HAADF-STEM images were recorded with drift-corrected frame integration (DCFI), which minimizes drift and achieves a high signal-to-noise ratio *via* calculating and correcting the drift from cross correlation.

RESULTS AND DISCUSSION

First, we used both X-ray diffraction (XRD) and SAED patterns of powder samples to check the phases of the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons. Some XRD peaks of the ribbons in Fig. 1h are quite similar to those of the DQCs of Al-Ni-Co [43], Al-Ni-Fe [43], and Al-Co-Cu [44], implying the possible existence of DQCs (also see the comparison of XRD in Fig. S1). This was further verified directly by the SAED patterns. The free side is Side A in Fig. 1a, and the side of the ribbon close to the copper roller is Side B. The grain sizes on Side B are evidently different from those on Side A because the cooling rate of Side A is lower than that of Side B. To determine whether the DQC is on Side A or B, we prepared TEM samples from both sides using an FIB instrument. We found that the DQC exists on both sides of the melt-spun ribbon. Fig. 1b, c show the bright-field TEM images of DQC particles from Side B viewed along the tenfold and twofold zone axes of the DQC, respectively. The DQC grains are on the nanoscale without an evident polygonal outline. Fig. 1d–f show the typical

Figure 1 Crystalline grains and DQC SAED patterns of the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbon. (a) Cross-sectional SEM image of melt-spun ribbon. (b, c) TEM images of quasicrystal grains. (d) SAED pattern along the tenfold zone axis from Side B. (e) SAED pattern of one twofold zone axis. (f) SAED pattern of another twofold zone axis at 18° from the SAED in (e). (g) SAED pattern along the tenfold zone axis of DQC in free side of the ribbon (Side A). (h) XRD patterns obtained from the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons.

SAED patterns of the DQC along the tenfold and two twofold zone axes from Side B. The diffraction spots in Fig. 1d show evident tenfold symmetry, implying it is a DQC [45]. Two representative twofold zone axes of the DQC in Fig. 1e, f are normal to the one in Fig. 1d, and each one contains periodic rows of diffraction spots (along the horizontal direction), corresponding to a periodicity of 1.23 nm in real space. Fig. 1g shows the typical SAED pattern along the tenfold zone axis of one DQC grain on the free side of the ribbon (Side A), where there is no obvious difference between Side A and Side B. The average DQC composition is determined to be

$\text{Al}_{20.5}\text{Si}_{19.0}\text{Mn}_{20.9}\text{Fe}_{26.3}\text{Ga}_{13.3}$, implying it is a multiple-principal-element DQC, different from any previously reported single-principal-element DQC [46]. The values of ΔS_{conf} for the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ alloy and the DQC phase are calculated to be $1.61R$ and $1.59R$, respectively. These are both greater than $1.5R$, thus satisfying the entropy-based definition of HEAs [16], which verifies that they are HEA and high-entropy phase, respectively. Note that the $\text{Al}_{20}\text{Si}_{20}\text{-Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ is a multiphase HEA, where crystal phases such as (1/0, 2/1)-type approximant of DQC [47], FeSi/MnSi-type (cubic, space group: $P2_13$ (198), PDF Card: 38-1397/42-1487) crystal are also found

in addition to the DQC. We do not present these crystals here because they are not the focus of this article.

To reveal the structural features of the high-entropy DQC, we obtained an atomic resolution HAADF-STEM image along the tenfold zone axis, as shown in Fig. 2. The structural blocks are depicted with sky-blue polygons by linking the centres of the smallest decagons with a diameter of ~ 0.47 nm. The most basic structural tiles with an edge length of 0.62 nm are squashed hexagon (H), star (S), boat (B), bowtie (BT), and decagon (D), as indicated at the bottom of Fig. 2a. We note that all of these basic structural blocks except D can further form several S tiles, shield-like tiles (SLTs) [48], concave decagons (CDs), and regular decagons, as seen clearly by filling different colours into the different types of tiles in Fig. 2b. Most of the

structural blocks in Fig. 2b are D tiles, along with several types (see in Fig. 3 below). The one containing three H tiles and one BT tile dominates (Fig. 2b), which is highlighted in light green. The D tiles can overlap with an area of one H tile or share one edge to generate a distance of 1.18 or 1.91 nm (nearly τ times the former, $\tau = 1.618$) between their centres. Fig. 2c shows the projected structural models of the basic structural blocks of BT, H, B, and S based on the atomic-scale HAADF-STEM image. The centres of the smallest D clusters are assigned to heavier elements such as Ga, Fe, and Mn because of the higher intensities in the HAADF-STEM image [49]. However, we cannot distinguish them owing to the difficulty in achieving atomic-resolution element mapping. Geometric configurations of different S, CD, and SLT

Figure 2 HAADF-STEM image and structural blocks of Al-Si-Mn-Fe-Ga DQC. (a) HAADF-STEM image along the tenfold axis of Al-Si-Mn-Fe-Ga DQC. (b) Colour-tiled HAADF-STEM image along the tenfold axis of Al-Si-Mn-Fe-Ga DQC. (c) Four basic structural blocks: BT, B, H, S. (d) Two types of S tiles. (e) Three types of concave decagon tiles. (f) Six types of SLTs.

kinds are schematically summarized in Fig. 2d–f. All types of each kind of tile have the same outline (highlighted in blue polygons). These tiles are composed of smaller tiles, therefore, owning the superiority over the smaller tiles in concision in describing the structural tiling [42].

The D clusters with a diameter of ~2.0 nm were considered the most important tiles in Al-based DQCs [7,42,46]. Therefore, in Fig. 3 we summarize the 2.0-nm D clusters in the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ DQC and propose the corresponding structural models. The common feature of these 2.0-nm D clusters is that the 0.47-nm D clusters are located at the periphery of the 2.0-nm D clusters. The tenfold symmetry of the 2.0-nm D cluster is broken gradually as the number of 0.47-nm clusters in the inner area increases, as seen from the left to right D clusters in Fig. 3. These structural features are quite similar to those of the $\text{Al}_{60}\text{Cr}_{20}\text{Fe}_{10}\text{Si}_{10}$ DQC [42] and different from other types of DQCs such as those in Al-Co-Ni, Al-Pd-Mn, and Al-Co-Cu alloys [46].

Phase prediction has long been a hot topic of discussion. The atomic size, crystal structure, valence electron concentration, and electronegativity of binary alloys were considered to have a major effect on phase formation according to the Hume-Rothery criterion [50], which was later amended by Zhang *et al.* [16,51] to extend to multi-

component alloys. The δ and ΔH_{mix} were initially used to predict phase formation in the multi-component alloys. Later, the effect of mixing entropy was also considered in phase formation, and then the supplementary criterion

$$\Omega = \frac{T_{\text{m}}^{\text{mix}} \Delta S_{\text{mix}}}{|\Delta H_{\text{mix}}|}, \text{ where } \Delta S_{\text{mix}} \text{ is the entropy of mixing, was}$$

proposed on the basis of the Gibbs free energy formula [51]. The solid solution phase usually forms when δ is smaller than 6.6%, ΔH_{mix} is between -15 and 5 kJ mol^{-1} , and Ω is greater than 1.1 [51–53]. However, the intermetallic and amorphous phases usually form at a larger δ and lower values of ΔH_{mix} and Ω [51–53].

Therefore, we calculated the ΔH_{mix} , δ , S_{conf}/R , and Ω values of the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ alloy and the DQC phase in this alloy, which are summarized in Table 1. After adding the points for $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ and the high-entropy DQC to the δ - ΔH_{mix} plot summarized by Ye *et al.* [53] (Fig. 4a), we found that neither the alloy nor the DQC phase belong to any classified areas. Combined with the fact that $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ is a multiphase HEA, the multiphase area in the δ - ΔH_{mix} diagram in [53] could be extended to include the current alloy, as seen in Fig. 4a. However, both the $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ alloy and the DQC phase therein are located in the amorphous area of the δ - Ω diagram by Yang *et al.* [51] (Fig. 4b). Considering the intermetallic feature of the DQC, we have

Figure 3 D clusters with the tenfold symmetry broken gradually. (a) Five types of D clusters. (b) Corresponding D clusters with the atoms superimposed. (c) Structural schematics of five types of D clusters. The tenfold symmetry of D clusters is broken gradually as the number of 0.47-nm D clusters increases inside (from left to right in the row of (a)).

Figure 4 Positions of $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ and $\text{Al}_{20.5}\text{Si}_{19.0}\text{Mn}_{20.9}\text{Fe}_{26.3}\text{Ga}_{13.3}$ in the δ - ΔH_{mix} (a) and δ - Ω (b) plots. We use stars to indicate the additional positions reported in this paper. (a) Adapted with the permission from Ref. [53], Copyright 2016, Elsevier; (b) adapted with the permission from Ref. [51], Copyright 2012, Elsevier.

Table 1 Calculated values of ΔH_{mix} (kJ mol^{-1}), δ , S_{conf}/R , and Ω

	ΔH_{mix} (kJ mol^{-1})	δ (%)	S_{conf}/R	Ω
$\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$	-25.60	7.67	1.61	0.65
DQC in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$	-26.08	7.13	1.59	0.68

extended the intermetallic area to include the DQC point in Fig. 4b. Therefore, the high-entropy quasicrystal alloy studied in this paper has expanded the area that is the criterion for HEA phases, and is complementary to the HEA-related δ - ΔH_{mix} and δ - Ω diagrams.

CONCLUSIONS

A near-equiatomic multi-component high-entropy DQC of $\text{Al}_{20.5}\text{Si}_{19.0}\text{Mn}_{20.9}\text{Fe}_{26.3}\text{Ga}_{13.3}$ was successfully synthesized in $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ melt-spun ribbons through a strategy using the structural similarity of different DQCs and the previously used factors of the δ and ΔH_{mix} . We studied the structural features of the high-entropy DQC along the tenfold axis in detail through HAADF-STEM imaging at atomic resolution. The most basic structural tiles of the high-entropy DQC are polygons with an edge length of 0.62 nm such as squashed hexagon, star, boat, bowtie, and decagon. We proposed the corresponding projected structural models on the basis of the HAADF-STEM imaging. Among these models, the decagons with a diameter of 2.0 nm have five types with their tenfold symmetry broken gradually as the number of 0.47-nm D clusters inside increases, which is analogous to the Al-based DQC in Al-Cr-Fe-Si alloys.

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Supplementary information Supporting data are available in the online version of the paper.

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$\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ 中近等原子比高熵十次准晶

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摘要 准晶是结构复杂相, 通常以一种合金元素为主要成分. 高熵合金含有多种主要合金元素, 其晶体结构却往往是比较简单的立方相. 作为结构和成分均复杂的高熵准晶却难以在实验上制备和理论上预测, 研究人员对其结构特点也知之甚少. 因而高熵准晶的制备和结构特性引起了人们的广泛关注. 我们报道了一种在 $\text{Al}_{20}\text{Si}_{20}\text{Mn}_{20}\text{Fe}_{20}\text{Ga}_{20}$ 甩带样品中发现的含有五个主要合金元素的近等原子比高熵十次准晶, 并对其晶体结构进行了详细表征. 该合金和十次准晶的构型熵均满足高熵合金基于熵的判据, 表明该十次准晶是高熵准晶. 该高熵十次准晶的设计策略为高熵准晶的制备提供了新的思路.