Microstructure and Mechanical Properties of FeCoNiCrMn High Entropy Alloy Reinforced Aluminum Matrix Composites Prepared by Friction Stir Processing

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(Received 30 September 2021; revised 31 August 2022; accepted 4 February 2023)

Abstract: High entropy alloy (HEA) particles are dispersed into 2024 aluminum alloy matrix by multi-pass friction stir processing (FSP) process to prepare HEA/Al composites. The effects of FSP passes on the macro morphology, microstructure and mechanical properties of the composites are studied. The microstructure observation shows that increasing FSP passes contributes to the uniform distribution of high entropy alloy particles and grain refinement. The test results of mechanical properties show that with the increase of FSP passes, the microhardness value of FSPed composites is significantly improved, the distribution is more uniform, and the strength and plasticity are also improved. The microhardness, strength and plasticity of five-pass FSPed composites reach 138 HV, 597 MPa and 5.1%, respectively.

Key words: friction stir processing; aluminum matrix composites; high entropy alloy (HEA); microstructure; mechanical property

CLC number: TG146; TG668 **Document code:** A **Article ID:** 1005-1120(2023)01-0098-08

0 Introduction

Since the 1930s, aluminum alloy has become the main material of choice for aircraft structural components because of its good design, manufacturing methods and high performance. 2024 Al alloy is an age hardening aluminum alloy, and copper is the main alloy element, which is mainly used for fuselage parts[1]. High entropy alloy is an alloy composed of five or more elements with equal molar ratio or near equal molar ratio. It inherits the characteristics of all elements and has excellent properties in strength, hardness and wear resistance, which makes it better than traditional alloys[2]. Aluminum matrix composites have attracted much attention because of their excellent mechanical properties and good application prospects, and are regarded as an ideal candidate for a new generation of lightweight high-strength materials[3]. Compared with aluminum alloy, aluminum matrix composites have higher Young's modulus, specific stiffness, specific strength and wear resistance[4-5], and have made certain applications in aerospace, transportation, electronic communication and other fields. Among them, particle reinforced aluminum matrix composites have always been one of the research hotspots in the field of metal matrix composites with the advantages of low cost, isotropy, and simple preparation process[6]. Due to the requirements of fuselage surface performance, several technologies have been developed, such as high-energy laser melting treatment[7-9], pulsed electron beam[5] plasma spraying[10-11], to prepare surface modified metal matrix composites (MMC). At present, the preparation

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How to cite this article: JIANG Jian, SHEN Yifu. Microstructure and mechanical properties of FeCoNiCrMn high entropy alloy reinforced aluminum matrix composites prepared by friction stir processing[J]. Transactions of Nanjing University of Aeronautics and Astronautics, 2023, 40(1): 98-105.

http://dx.doi.org/10.16356/j.1005-1120.2023.01.009

technologies for particle reinforced aluminum matrix composites include stirring casting, powder metallurgy, and spray molding, but these technologies are often carried out at high temperature, and the chemical reaction and brittle phase will occur at the interface between particles and matrix. Therefore, it is urgent to find a new preparation technology. Friction stir processing (FSP) is a new surface modification technology based on the principle of friction stir welding (FSW)[12-14]. FSW is a solid-state connection method, which is mainly applied to aluminum alloy, and then applied to other nonweldable light alloys, such as copper, nickel, and titanium. This technology needs simple process and low energy consumption. It is a green advanced manufacturing technology[15-18]. Therefore, in this paper, FeCoNiCrMn high entropy alloy particles are used as the reinforcing phase and 2024 aluminum alloy is used as the matrix to prepare high entropy alloy particle reinforced aluminum matrix composites by FSP technology. The effects of processing pass on the appearance, microstructure, microhardness and tensile properties of 2024 aluminum matrix composites are studied.

1 Materials and Methods

The size of 2024 aluminum alloy used in the experiment is 150 mm × 150 mm and the thickness is 5 mm. The composition is shown in Table 1. The powder used in this study is commercial FeCoNiCrMn powder. Its composition is shown in Table 2, the particle morphology is shown in Fig.1(a) and its XRD pattern is shown in Fig.1(b). FeCoNiCrMn high entropy alloy is mainly composed of face centered cubic (FCC) single-phase solid solution.

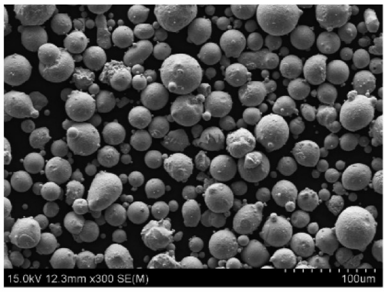
Table 1 Chemical composition of 2024 Al alloy (weight percent)

Element	Cu	Mg	Mn	Fe	Si	Zn	Ti	Al
Content/%	4.6	1.52	1.45	0.15	0.06	0.08	0.048	Bal.

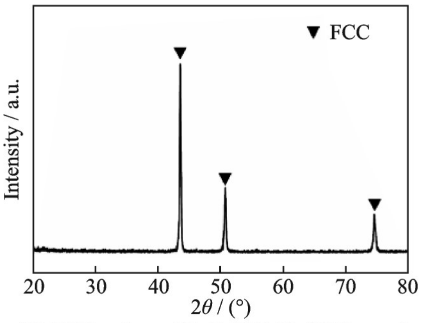
In order to add HEA powder to the aluminum plate, a groove with a depth of 2.0 mm and a width of 1.5 mm is machined along the longitudinal direction of the aluminum plate, and the tungsten powder is put into the groove for compression. The workpiece with pre-placed HEA powder is clamped on the test platform to fix the sample in place during FSP process. In order to avoid the loss of HEA particles from the groove during FSP process, a pin-free stirring head is used to close the groove opening in advance. The numerical control milling machine is used for FSP. The stirring pin is inserted into the aluminum alloy surface and moves along the center line of the groove. For multi pass FSP, the stirring head moves back and forth along the same line, and the next FSP is carried out after the workpiece is cooled from the previous pass to room temperature. The FSP process and process parameters for preparing HEA reinforced aluminum matrix composites are shown in Fig.2 and Table 3, respectively.

Table 2 Chemical composition of high entropy alloy reinforcement phase (weight percent)

Element	N	O	Fe	Co	Cr	Mn	Ni
Content/%	0.013 7	0.041 0	19.55	21.18	18.84	19.88	Bal.



(a) Morphology of FeCoNiCrMn HEA powder



(b) XRD pattern of FeCoNiCrMn HEA powder

Fig.1 Morphology and XRD pattern of FeCoNiCrMn HEA powder

After FSP, the cross section of the FSPed sample perpendicular to the travel direction is cut, polished and corroded with Keller reagent (1.0 ml HF, 1.5 ml HCl and 2.5 ml HNO_3 dissolved in 95

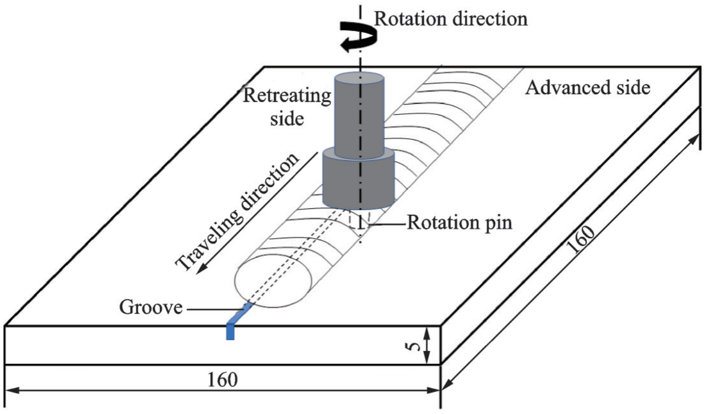


Fig.2 Schematic diagram of FSP

Table 3 Process parameters of FSP

Sample	Rotation speed/ (r•min^{-1})	Traveling speed/ (mm•min^{-1})	Plunge depth/ mm	FSP pass
1	800	60	0.20	1
2	800	60	0.30	3
3	800	60	0.40	5

ml distilled water). The microstructure evolution from base metal to stirring zone under different FSP passes are studied by optical microscope (OM) and field emission scanning electron microscope (FESEM).

WDW-1000 universal tensile testing machine is used to evaluate the mechanical properties of machined samples of surface composites under the condition of initial strain rate of $1 \times 10^{-3} \mathrm{s}^{-1}$. The tensile properties are compared with 2024 aluminum alloy. The tensile specimen is prepared according to ASTM E8/E8M-11 and processed to the required size with a wire cutting machine. The sample size is shown in Fig.3, and the thickness of the tensile sample is 2.0 mm. After the tensile test, the fracture is analyzed by scanning electron microscope (SEM) to reveal the influence of FSP passes on the fracture behavior. HXS-1000 hardness tester is used to measure the microhardness along the cross section of the nugget zone of the processed sample. The load is 200g and the pressure dwell time is 15 s.

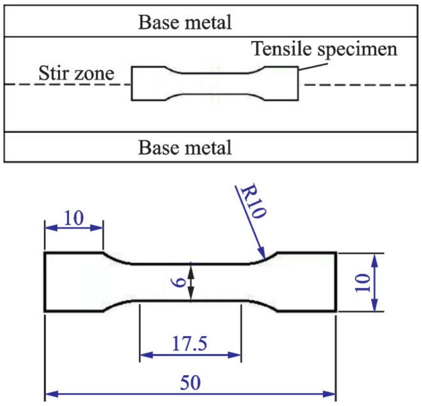
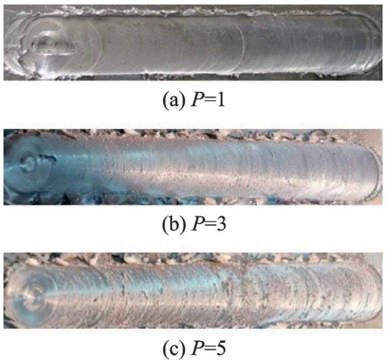


Fig.3 Schematic diagram of cutting position and size of tensile specimen

2 Results and Discussion

The surface macro morphology of 2024 aluminum alloy plate after different passes of FSP is shown in Fig.4. During single pass ($P = 1$) processing, the surface of the processing area is smooth with a small amount of flash at the interface of the processing area and large tunnel defects in the weld. When the processing pass is 3, the tunnel defects in the processing area are alleviated and the surface quality is also reduced. After the number of processing pass increases to 5, the flash at the interface between the processing area and the base metal area increases. This is because the lower pressure of the shaft shoulder in the later pass is higher than that in the previous pass, resulting in some metals extruding the shoulder and forming flash. Although there are no obvious defects, the finish decreases slightly, the roughness increases slightly, and the flash adhesion in multiple passes leads to the decline of surface quality. Therefore, after multi-pass FSP, the defects in the nugget zone are reduced, but the surface appearance quality is slightly reduced. In the process of FSP, when the processing pass is one,


(a) P=1

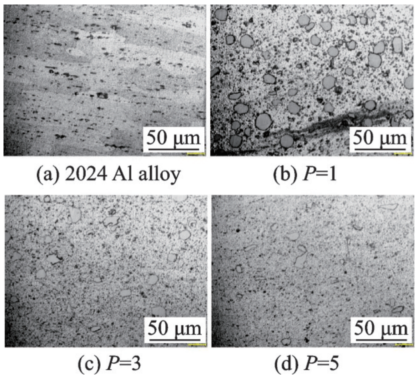
(b) P=3

(c) P=5

Fig.4 Weld morphology of different FSP passes

because the heat generation of the shoulder is far greater than that of the top of the mixing pin, the plasticization degree of the upper material in the processing area is greater than that of the lower material. Thus, the material fluidity in the upper part of the processing area is better, while the fluidity in the lower part is poor, and there is material extrusion in the processing area, eventually leading to the formation of tunnel defects. When the number of processing passes increases, the heat production in the lower part of the processing area increases relatively, and the phenomenon of low degree of material plasticization is improved, improving the material fluidity of the whole processing area and avoiding the generation of tunnel defects.

The metallographic structure of 2024 aluminum alloy base metal and mixing zone of aluminum matrix composites in different processing passes is shown in Fig.5. Fig.5 shows that, when processing a single pass, the size of the second phase of HEA particles in the stirring zone is large, the distribution is uneven, the particle segregation is serious, and the particle fragmentation is not obvious. The particle size of the second phase with three passes is significantly lower than that with a single pass, and the length is more than 30 μm. The large-size particles are greatly reduced. The improvement of segregation is also obvious. With the further increase of processing passes, i.e. $P = 5$, the refinement effect of the second phase particles is further improved. Compared with the sample of a single pass, the proportion of ultra-fine particles is significantly increased, and the distribution of the HEA particles in 2024 Al matrix is more uniform.



(a) 2024 Al alloy

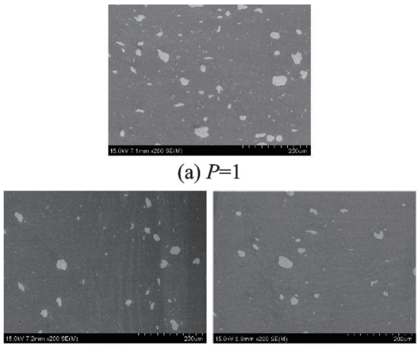
(b) P=1

(c) P=3

(d) P=5

Fig.5 Optical microstructures of 2024 aluminum alloy and aluminum matrix composites with different passes of FSP

Fig.6 shows SEM micrographs of FSPed composites prepared under different processing passes. It can be clearly seen from Fig.6(a) that there are obvious HEA clusters in the single pass FSPed composite. This is mainly due to the following two reasons: (1) When the HEA particles are pressed into the groove, the pressure causes the HEA particles to be mechanically interlocked and cold welded together; (2) large residual strain and residual stress are produced in the production process of hot-rolled aluminum plate, which leads to insufficient material softening and poor plastic flow during one-time rolling. With the increase the pass of FSP, the particle size of HEA decreases significantly. As shown in Fig.6(b), only a small amount of HEA particle segregation is observed in the three-pass FSPed composite, while a more uniform HEA particle distribution is obtained in the five-pass FSPed composite, and there is almost no agglomeration of HEA particles. This is because the accumulated plastic strain and repeated heat input lead to greater plastic deformation and thorough mixing. However, with the increase of FSP pass, the size and morphology of HEA particles change significantly, and the particle size decreases significantly. This observation is consistent with the results reported by Prater[19] who reports that the violent stirring effect and strong plastic strain of the stirring head will destroy the ceramic particles and change their size and morphology. In


(a) P=1

(b) P=3

(c) P=5

Fig.6 Microstructure of stir zone (SZ) under different processing passes

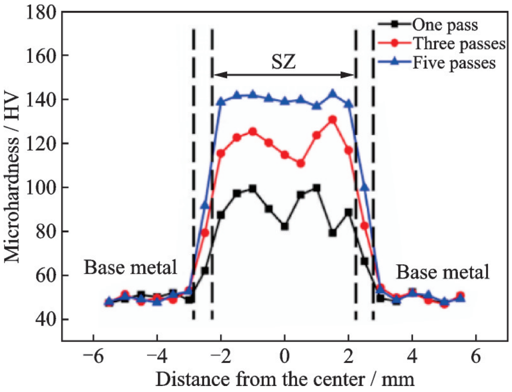


Fig.7 Microhardness distribution of HEA/Al composites by different passes of FSP

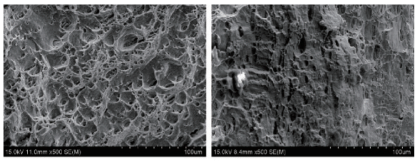
addition, it is worth noting that SEM micrographs of all composites show no particle shedding during mechanical polishing. This may mean that effective HEA/Al interface bonding is obtained. It also shows the effectiveness of five-pass FSP process for the preparation of composites with uniform microstructure. The HEA/Al interface has very good interface integrity and no micropores. This can be attributed to the fact that Al with high plasticity can wet the whole surface of HEA particles, so as to avoid the formation of micropores and undercuts at the HEA/Al interface. Obviously, good particle/matrix interface bonding helps to transfer the load from the matrix to the reinforced particles through the particle/matrix interface. In addition, the SEM photos of all FSPed composites, even in the case of five passes, show that the HEA/Al interface is clean and there is no obvious sign of reaction layer formation. It is generally believed that the lack of brittle intermetallic compounds plays an important role in improving the interfacial properties and the ductility and strength of composites.

Fig.7 shows the microhardness values of FSPed composites with different passes. It can be seen that the microhardness of FSPed composite is much higher than that of base metal (approximately 48 HV). Especially with the increase of FSP pass, the microhardness continues to increase. When the processing pass is five, the average microhardness of FSPed composite reaches the maximum of 138 HV. The increase of microhardness can be attributed to the following three reasons: (1) Orowan mechanism due to the existence of HEA reinforcing phase[20-21]; (2) Hall-Petch strengthening mechanism caused by fine grain of Al matrix; (3) thermal expansion dislocation strengthening caused by dislocations due to different thermal shrinkage coefficients of HEA particles and Al matrix[22]. In addition, it should be noted that increasing FSP pass can make the microhardness distribution more uniform. This can be attributed to the better dispersion of HEA particles in higher FSP passes.

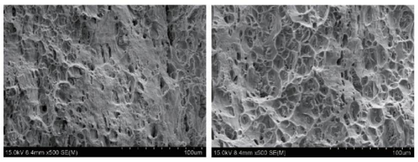
Tensile tests are carried out on 2024 aluminum alloy base metal and FSPed aluminum matrix composites with different passes. The test results are shown in Table 4. Friction stir machining can effectively improve the yield strength and ultimate tensile strength (UTS) of aluminum alloy, and its yield strength and UTS gradually increase with the increase of machining pass. The elongation of aluminum matrix composites prepared by friction stir processing is lower than that of aluminum alloy base metal, but the plasticity of aluminum matrix composites can be significantly improved with the increase of processing pass. Fig.8 shows SEM images of tensile fracture of 2024 aluminum alloy base metal and FSPed aluminum matrix composites with different passes. As shown in Fig.8(a), a large number of deep dimples and shear ridges are observed on the fracture surface of 2024 aluminum alloy, which indicates that the fracture is a typical ductile fracture, and its failure mechanism is the nucleation and consolidation of holes. Figs.8(b—d) show the tensile fracture of FSPed composites with different passes. As shown in Figs.8(b, c), for one-pass and three-pass FSPed composites, HEA clusters and dispersed HEA particles can be observed on the fracture, which is consistent with the distribution of HEA shown in Figs.5(b, c). According to the flat fracture surface in the HEA cluster area, it can be inferred that the poor plasticity of low FSP passes may be related to HEA clusters, because HEA clusters will lead to stress concentration, which will induce microporous nucleation in the nearby Al matrix and reduce the plasticity concequently. On the fracture morphology of the five-pass FSPed composite shown in Fig. 8(d), the uniform distribution of pulled out HEA particles without HEA clusters can be observed in the dimple, which is consistent with the HEA distribution shown in Fig.6(c). Comparing the dimples in the fracture of base metal and aluminum matrix composites shows that HEA particles have a limiting effect on dislocation mobility, which confirms that there is a good interfacial bonding between aluminum matrix and HEA particles. Therefore, five-pass FSPed composites have high strength and good plasticity.

Table 4 Mechanical properties of 2024 Al alloy and Al matrix composites with different processing passes

Sample	0.2% proof stress/ MPa	UTS /MPa	Elongation /%
Base metal	374	439	6.3
$P=1$	399	477	3.1
$P=3$	465	532	3.8
$P=5$	518	597	5.1



(a) 2024 Al alloy (b) P=1



(c) P=3 (d) P=5

Fig.8 SEM images of tensile fracture of 2024 aluminum alloy base metal and HEA/Al composites by one-pass, three-pass and five-pass FSPed composites

3 Conclusions

HEA/Al composites are prepared by multi-pass friction stir process. The microstructure, HEA/Al interface bonding, composition, microhardness and tensile properties of different FSPed composites are studied. From the results obtained, the following conclusions can be drawn:

(1) Due to the accumulated plastic strain and repeated heat input of multi-pass FSP, the size of HEA particles is reduced and the distribution is more uniform.

(2) There are no intermetallic compounds and oxides in five-pass FSPed HEA/Al composites. The five-pass FSPed composite forms a good metallurgical bonding at the HEA/Al interface.

(3) Increasing FSP pass is helpful to improve the microhardness and its uniform distribution. The maximum microhardness of aluminum matrix composites obtained by five passes of FSP is 138 HV, which is 2.9 times that of 2024 aluminum alloy.

(4) The strength and plasticity of FSPed aluminum matrix composites increase with the increase of FSP pass. The maximum tensile strength and elongation of five passes of FSP reach about 597 MPa and 5.1%, respectively.

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Acknowledgements This work was a project funded by the Priority Academic Program Development of Jiangsu Higher Education Institution. This research is also supported by Beijing Institute of Aeronautical Materials (No.KZ82171509) and China Scholarship Council (CSC).

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Author contributions Mr. **JIANG Jian** manufactured the samples, performed the experiments, and wrote the manuscript. Prof. **SHEN Yifu** designed the study, reviewed the manuscript and contributed to the background of the study. Both authors commented on the manuscript draft and approved the submission.

Competing interests The authors declare no competing interests.

(Production Editor: XU Chengting)

搅拌摩擦加工制备FeCoNiCrMn高熵合金增强铝基复合材料的组织与力学性能研究

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摘要：采用多道搅拌摩擦加工（Friction stir processing, FSP）工艺将高熵合金（High entropy alloy, HEA）颗粒分散到2024铝合金基体中，制备了HEA/Al复合材料。研究了搅拌摩擦加工道次对复合材料宏观形貌、微观结构和力学性能的影响。显微组织观察表明，增加搅拌摩擦加工道次有助于高熵合金颗粒的均匀分布和晶粒细化。力学性能测试结果表明，随着搅拌摩擦加工道次的增加，搅拌摩擦加工制备复合材料的显微硬度值显著提高，分布更加均匀，强度和塑性也有所提高。五道次搅拌摩擦加工所制备复合材料的显微硬度、强度和塑性分别达到138 HV、597 MPa和5.1%。

关键词：搅拌摩擦加工；铝基复合材料；高熵合金；显微组织；机械性能