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Synthesis and characterization of a new high entropy composite matrix

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Abstract. Even if high entropy alloys were not reported in a scientific journal till 2003, these new alloys have been investigated since 1995 due to their high temperature properties. In the last years the synthesis of these alloys has been widely investigated. Thus, the present work has been carried out to produce a high entropy composite using an equiatomic AlCrFeMnNi high entropy alloy (HEA) matrix and graphite particles (Gr) as reinforcing material. The high entropy composite was obtained by powder metallurgy route using a planetary ball mill. The mechanically alloyed mixture was investigated by scanning electron microscopy (SEM). Microstructural investigation realized by SEM revealed the homogenous structure of the composite, with multiple phases and decreasing particles size, mostly reaching nanometric scale.

1. Introduction

Conventional alloys are based on one principal element with different elements added to improve their properties [1]. Based on the existing research and literature on classical alloys, Jien-Wei Yeh explored the multicomponent world and after years of research he developed the HEA concept providing experimental results and related theory. He defined HEAs as those alloys containing at least 5 principal elements with the atomic percentage of between 5% and 35%, with the atomic percentage of each minor element, if any, less than 5% [2].

Due to the combinations of composition and process for producing HEAs and each HEA having a particular microstructure and properties to be identified and understood, the research work is almost limitless [3, 4]. HEAs have shown interesting properties such as high hardness and high strength, good thermal stability wear and oxidation resistance, properties which provide great potential for engineering applications [4 – 9]. The high entropy alloys have been produced by different routes, the usual route being liquid processing.

During the last years, mechanical alloying technique has been more often approached due to the good homogenous chemical distribution and near net shape technology provided by the respectively process [9, 10, 11]. Chao Wang, Wei Ji and Zhengyi Fu obtained simple structured solid solution after 30 h of milling, with a grain size of less than 20 nm while B.S.Murty et al. succeeded to obtain a HEA with BCC structure and grain size of approximately 10 nm [11,12,13].

This report aims to reveal the synthesis method for a composite material using a HEA matrix and Gr particles as reinforcing material. Based on this objective, an equiatomic AlCrFeMnNi HEA matrix was fabricated by MA process and Gr was added after the HEA was obtained. The alloy microstructure was investigated and the XDR analyses were performed to see the moment when the mechanical alloying was produced.

2. Materials and Methods

Al, Cr, Fe, Mn, Ni elemental powders of >99% purity and ≤45 mμ, used as matrix, were mechanically alloyed in a high energy planetary ball mill (Retsch 400 PM) for 40 h in Ar atmosphere at 300 rpm with a ball to powder ratio (BPR) of 10:1. Graphite was added to obtain a new HEA composite. The graphite particles were added 3% (volumetric) to improve the powder flow and weldability. Stainless steel vials and balls have been used. Heptane as process control agent (PCA) was used to prevent powder deposition on the vials walls and milling balls. Samples were extracted at intervals of 20, 25, and 40 hours.

The mechanically alloyed powder was microstructural characterized. QUANTA INSPECT F scanning electron microscope (SEM) foreseen with a field emission electron gun, and equipped with analysis dispersive system in X-radiation energy was used to observe the morphology and particle reduction. The samples were milled at different times in order to assess the best milling time for a planetary ball mill, with 300 rpm, 10:1BPR to process a high entropy composite material. Samples taken at different intervals revealed that after 20 hours the alloying process began.

3. Results and discussions

Figure 1 reveals the AlCrFeMnNIGr HEA composite microstructure under different milling times.

The particles in the primitive powder are roughly between 5 and 45 μm and of irregular shape, with sharp edges, as seen In Figure1 (a). The ball to powder ratio chosen was 10:1 and the ball mill speed was 300rpm. The idea of the experiment was to assess the milling time necessary for the alloying process to be produced. The authors tried to decrease as much as possible the milling time using a planetary ball mill with an optimum speed of 300rpm. Previously the milling time reported was of 60 hours [9] but with a BPR of 7:1. With the increase of the milling time the particles have reduced their size exponentially with time and cold weld together to form agglomerations of elliptical shape after 20 and 25 h as seen in figure 1 (b) and (c). Figure 1 (d) reveals that at 40 h milling time further decrease in particles size with grain size reaching nanometric scale as it will be further described by the diffraction pattern. The microstructure of the 20 h milled sample shows a good homogeneity and the welding process of the particles began.

The powder further processed present agglomeration for the particles of different powder. The alloyed particles have a light grey colour, uniform and the particles are so small that the agglomeration could not be avoided. The percentage of graphite particles added improved the milling process and the powder was alloyed after 40 hours milling. In further papers the properties of the new composite material obtained will be investigated. The graphite particles were added from the beginning to obtain a homogeneous structure.

The microstructure study is important for revealing the sample homogeneity. To establish if the component of the powder were alloyed after the process an EDS analysis was performed.

In figure 2 EDS analyses results show the homogeneity of the mentioned HEA composite during the mechanically alloying process. In figure 2a the distribution of the particles reveals all elemental powder used to create the high entropy composite. Figure 2b shows a mixture of all these elements showing that the alloying process began.

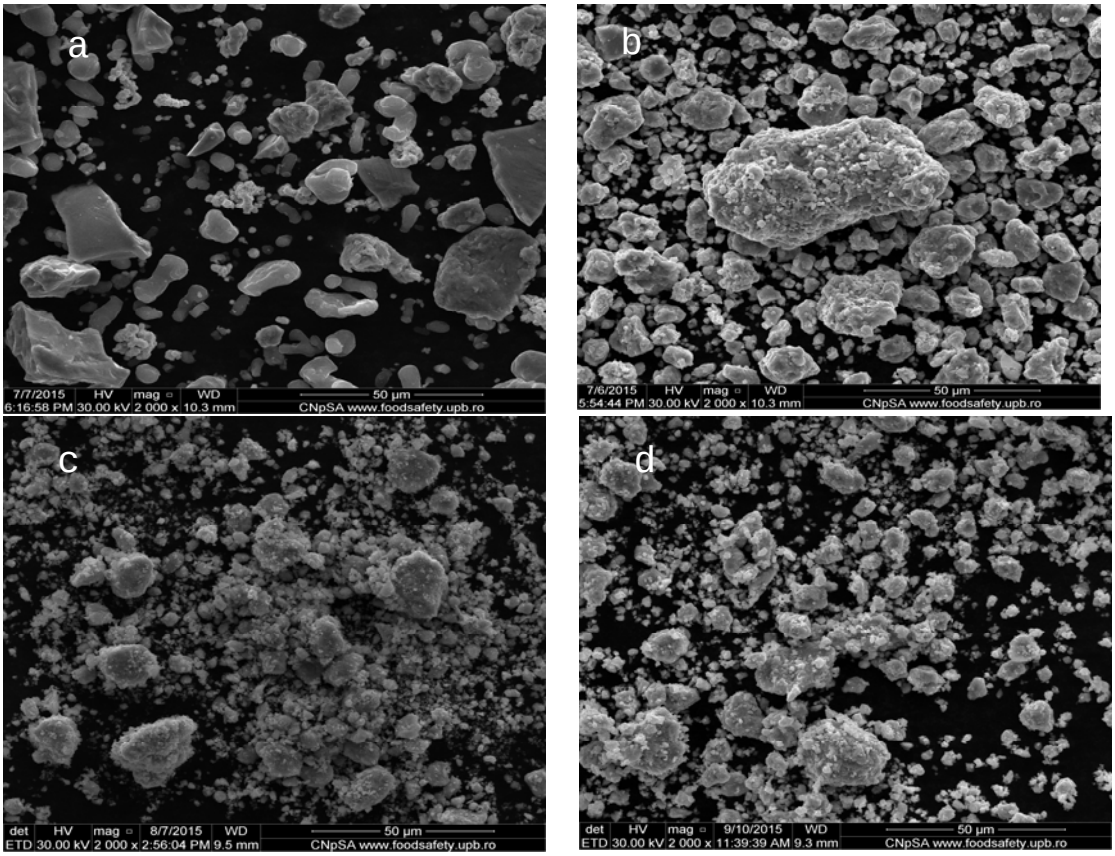

Figure 1. SEM image of AlCrFeMnNiGr HEA composite at (a) 0 h; (b) 20 h; (c) 25 h; (d) 40 h.

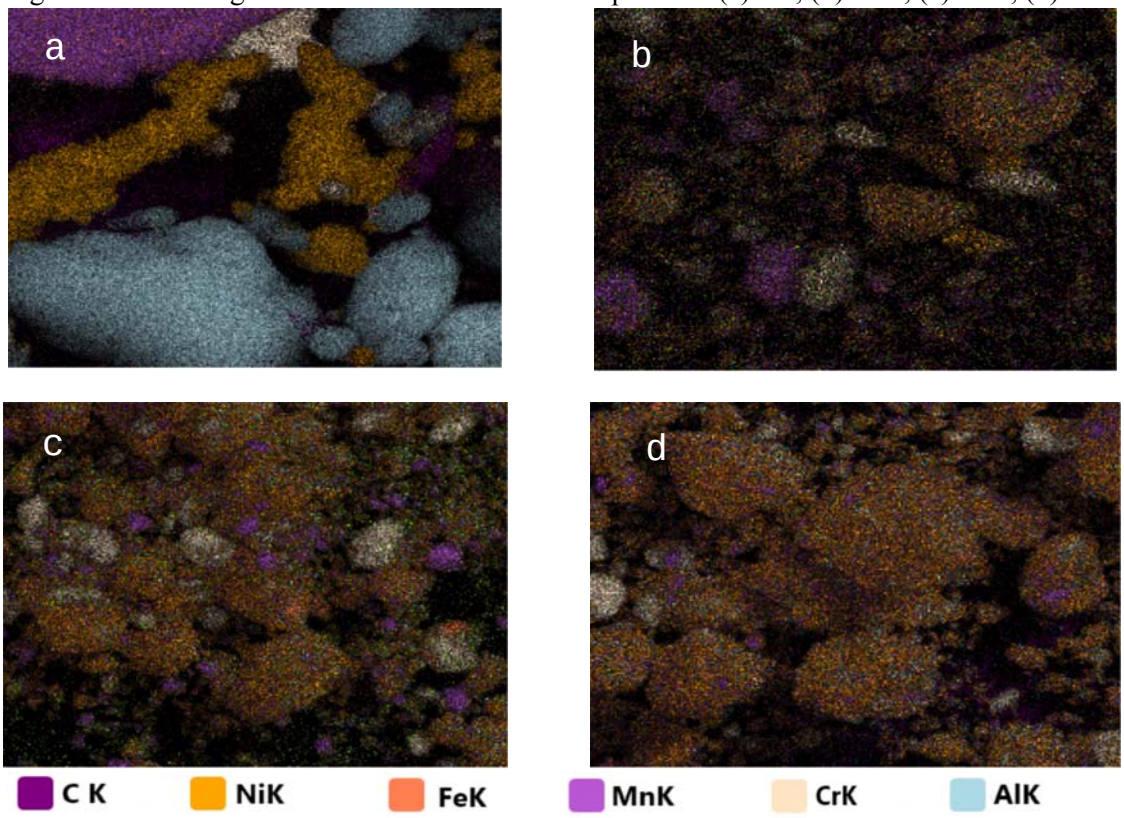

Figure 2. EDS of AlCrFeMnNiGr HEA composite: a) 0 h, b) 20 h, c) 25 h, d) 40 h.

The authors assessed the microstructure of the samples after 25 hours (figure 2c). The EDS analyses revealed the alloying process in a better phase than after 20 hours but still Mn particles could be observed unalloyed. After 40 h milling time a homogenous structure can be observed with the elements evenly distributed.

Figure 3 shows the XRD pattern of AlCrFeMnNiGr HEA composite before and after milling time. The peaks of all the pure elements in the powder before alloying could be observed in figure 3, in the initial mixture. After 40 h of milling time most of the peaks disappear revealing the solid solution formation. The transformation of the phases from FCC to BCC can be observed in the diffraction pattern. The blue line indicates also the absence of the other elemental peaks proving the fact that the alloying process was realized.

Table 1 exhibits crystallite size and lattice strain for 40 h milling time, calculated with Scherrer's formula

$$D = (K\lambda)/(\beta\cos\theta) \quad (1)$$

Where λ is the X-ray wavelength in nanometres (nm), β is diffraction broadening and K is a constant. The θ can be in degrees or radians [14]. The Scherrer formula was used to determine the crystallite size and lattice strain which is one of the four core effects of HEAs. Because of the atomic size difference lattice strain occurs increasing hardness and strength but also giving a higher resistance of the material properties at high temperatures [2].

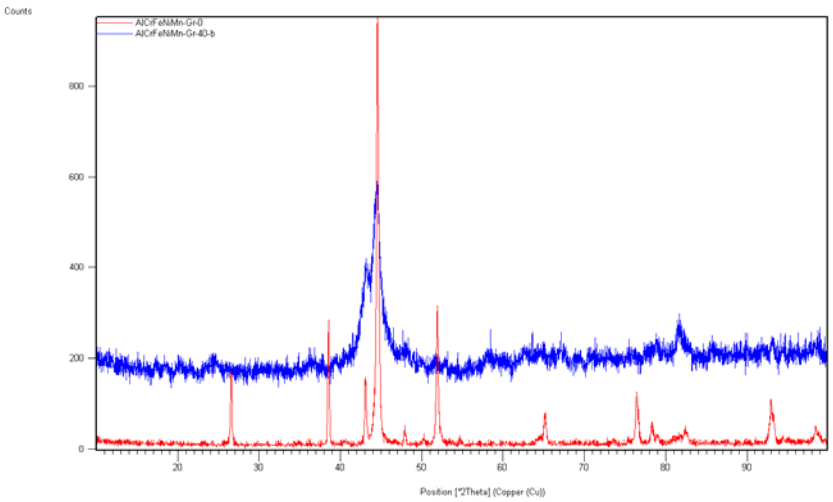

Figure 3. XRD patterns at 0 and 40 h milling time of AlCrFeMnNiGr HEA composite.

Table 1. The crystallite size and lattice strain of AlCrFeMnNiGr HEA at 0 and 40 h milling time

Milling time/h	Crystallite size/nm	Lattice strain/%
40	39.28	2.59

4. Conclusions

A high entropy alloy was syntheses starting from the pure elemental powders from Al, Cr, Fe, Ni, Mn. To enhance this high entropy alloy properties graphite particles were added as reinforcing elements. The graphite particles were added from the beginning to improve the powders properties.

The high entropy composite was alloyed for 20, 25 and 40 hours. The microstructure studies revealed that the composite obtained was homogeneous and EDS analyses results showed that the elements are uniformly distributed.

The XRD analysis revealed the solid solution formation and the nanometric size of the particles.

The high entropy composite was successfully synthetized after 40 hours of milling time.

The lattice strain calculate with the aid of Scherrer formula shows a value characteristic for high entropy alloys, providing a better behaviour for these type of alloys at elevated temperatures.

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