Arch. Metall. Mater. **64** (2019), 3, 949-952

DOI: 10.24425/amm.2019.129478

JU-YEON HAN*, HYUNJI KANG*, SUNG-TAG OH*#

EFFECT OF PROCESS CONTROL AGENT ON THE MICROSTRUCTURE OF Ni-BASED ODS SUPERALLOY PRODUCED BY MECHANICAL ALLOYING AND SINTERING

The effects of different types of process control agents (PCA) on the microstructure evolution of Ni-based oxide dispersion-strengthened superalloy have been investigated. Alloy synthesis was performed on elemental powders having a nominal composition of Ni-15Cr-4.5Al-4W-2.5Ti-2Mo-2Ta-0.15Zr-1.1Y_2O_3 in wt % using high energy ball milling for 5 h. The prepared powders are consolidated by spark plasma sintering at 1000°C. Results indicated that the powder ball-milled with ethanol as PCA showed large particle size, low carbon content and homogeneous distribution of elemental powders compared with the powder by stearic acid. The sintered alloy prepared by ethanol as PCA exhibited a homogeneous microstructure with fine precipitates at the grain boundaries. The microstructural characteristics have been discussed on the basis of function of the PCA.

Keywords: Ni-based superalloy, Process control agents, Mechanical alloying, Spark plasma sintering, Microstructure

1. Introduction

Ni-based oxide dispersion strengthened (ODS) superalloys are promising materials for aircraft and gas turbine applications due to their excellent strength and high creep resistance [1,2]. They achieve excellent mechanical properties at high temperature by combination of gamma prime precipitation strengthening and oxide dispersion strengthening [3,4]. Generally, fabrication of ODS superalloys involves the mechanical alloying (MA) process and consolidation by heat treatment. The MA process is the core step to synthesis homogeneous superalloy powders, in which the elemental metal and oxide powders are subjected to the high energy ball milling to allow the oxide particles to be finely and homogeneously dispersed in the alloy matrix [1,5].

However, in order to develop the desired final chemical composition, homogeneity and grain refinement, the optimization of milling process such as the avoidance of excessive cold welding and contamination is needed [6,7]. Nevertheless, the microstructure evolution of Ni-based ODS superalloy powders dependent on the process-control agents (PCA) has not been comprehensively reported [8]. This study aims to analyze the effect of different types of PCA, i.e., stearic acid and ethanol, on the microstructure of ball-milled powder and sintered specimen having a nominal composition of Ni-15Cr-4.5Al-4W-2.5Ti-2Mo-2Ta-0.15Zr-1.1Y_2O_3 in wt %. Also, an optimum synthesis condition is suggested based on the observed microstructural characteristics of Ni-based ODS superalloy.

2. Experimental

Ni-based ODS superalloy powders were prepared using elemental Ni (8 μm), Cr (28 μm), Al (18 μm), W (12 μm), Ti (26 μm), Mo (3-10 μm), Ta (2 μm), Zr (2-15 μm) and Y_2O_3 (0.05 μm) powders. The starting powders were initially mechanically mixed in a tubular mixer for 1 h. Typical morphology of mixed powder is shown in Fig. 1. The mixed powders were then placed into stainless steel vessel with balls that were sealed and filled with Ar gas. The MA was carried out for 5 h using hori-

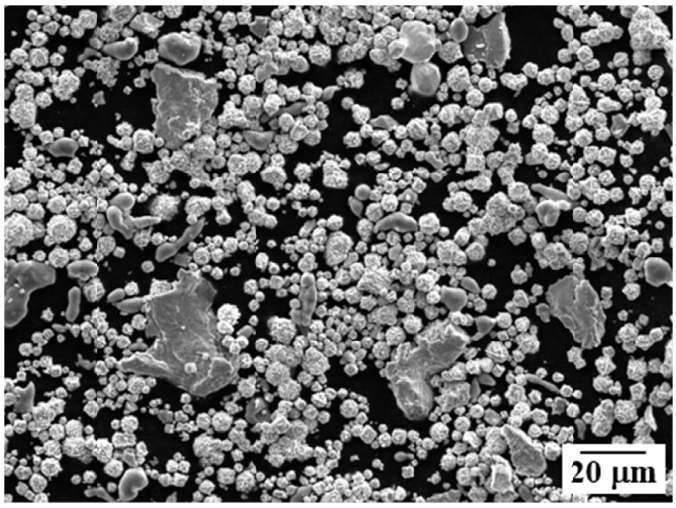

Fig. 1. SEM image of mixed powders in a tubular mixer for 1 h

* SEOUL NATIONAL UNIVERSITY OF SCIENCE AND TECHNOLOGY, DEPARTMENT OF MATERIALS SCIENCE AND ENGINEERING, SEOUL 01811, REPUBLIC OF KOREA

Corresponding author: stoh@seoultech.ac.kr

zontal ball milling machine (Simoloyer, Zoz GmbH, Germany) by cyclic operation of milling velocity with a ball-to-powder weight ratio of 15:1 [9]. In order to investigate the effects of the species of PCA on the MA process, 1 wt% of stearic acid ($CH_3(CH_2)_{16}COOH$) and ethanol (C_2H_5OH) was added to the total elemental powders, respectively. The PCA was removed by heating the ball-milled powders in air at 400°C for 2 h. The MA powders were sintered at 1000°C for 10 min in vacuum under a pressure of 30 MPa using spark plasma sintering (SPS, Sumitomo Coal Mining, Japan).

The size of ball-milled powders was measured by laser particle size analyzer (LS I3 320, Beckman Counter, USA). The carbon content was analyzed by automatic elemental analyzer (Flash EA1112, Thermo Finnigan). The processed powder and sintered specimen were characterized by X-ray diffraction (XRD, D/Max-IIIC, Rigaku Denki Co., Japan) analysis. The microstructure and elemental mapping were observed by scanning electron microscopy (SEM, JSM-6700F, JEOL Co., Japan) equipped with an electron probe micro-analyzer (EPMA).

3. Results and discussion

Particle size analysis results of ball-milled powders with the addition of 1 wt% stearic acid and ethanol are shown in Fig. 2(a) and (b), respectively. It can be seen that the ball-milled powders with the addition of stearic acid exhibited smaller particle size and more uniform distribution of particle size than that with ethanol. The measured mean size and carbon content in the powder mixtures are given in Table 1. In case of milling in stearic acid as a PCA, the mean particle size and carbon content were 13.2 μm and 0.11 wt%, while the powder mixture milled in ethanol showed the value of 27.6 μm and 0.02 wt%. These results indicated that the use of stearic acid can significantly decrease the particle size by the rapid fracturing of cold-welded powders and effective reduction of fresh surface contacting [10].

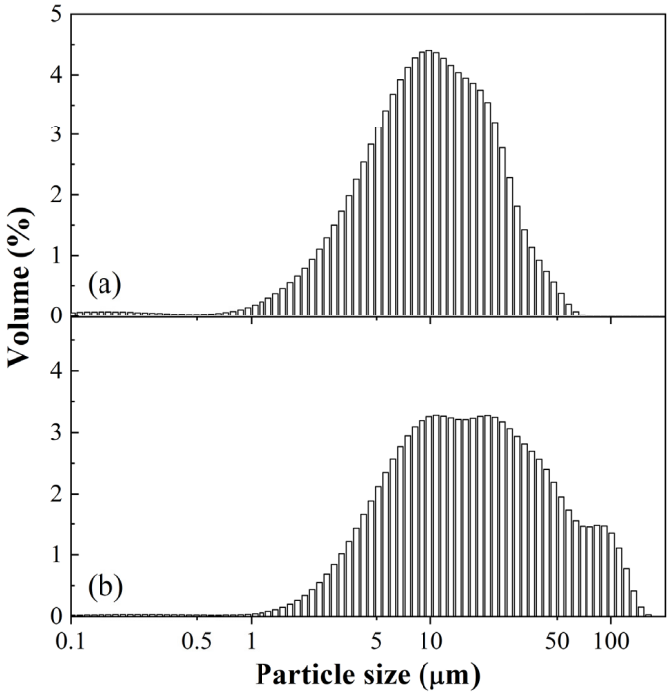

Fig. 2. Particle size distribution of the ball-milled powders with 1 wt% of (a) stearic acid and (b) ethanol as PCA

TABLE 1

Particle size distribution and carbon content of ball-milled powders by using different PCAs

PCA	Mean particle size (μm)	Carbon content (wt%)
Stearic acid	13.2±10.4	0.11
Ethanol	27.6±28.4	0.02

The distribution of alloy elements in the ball-milled powder mixtures is characterized by SEM and EPMA mapping. As clearly seen in Fig. 3(a) and (b), although the size of ball-milled powders with stearic acid was small, it represented a more uni-

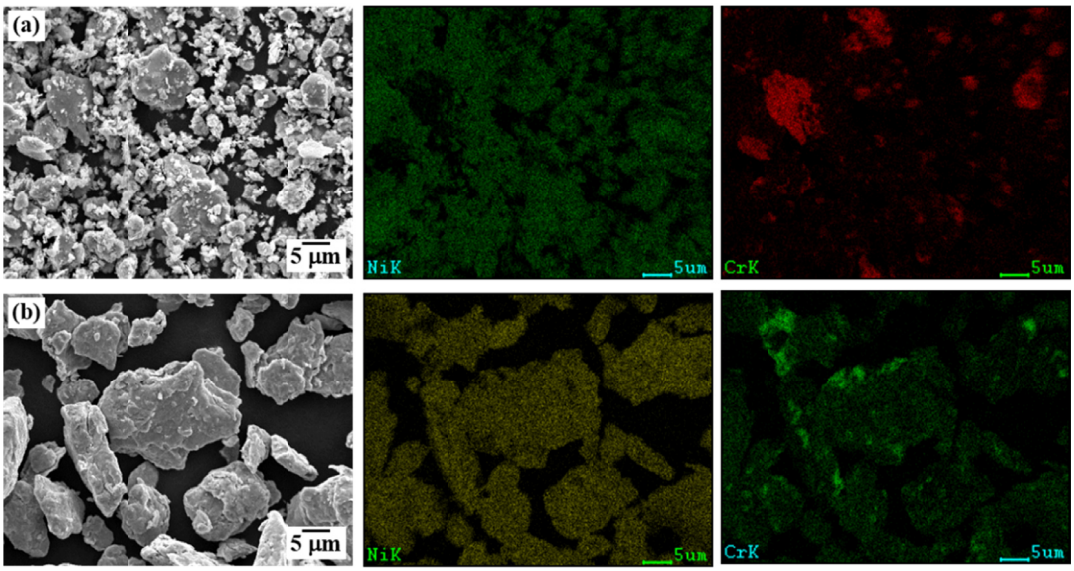

Fig. 3. SEM images and related EPMA maps of ball-milled alloy powders with (a) stearic acid and (b) ethanol

form distribution of main elements in the powder prepared by ethanol as PCA. The reason for the different effect of stearic acid and ethanol on ball-milled powders is not fully understood yet. But possible mechanisms are proposed in the literatures [11,12] that the change of particle size and distribution of alloy elements with different types of PCA is attributable to the different melting point and molecule size of PCA.

Fig. 4 shows the XRD profiles of the Ni-based ODS superalloy powders at different stages of processing. The ball-milled powder for 5 h shows the peaks of all major alloying elements such as Ni, Cr, Al and W as well as Al_2O_3 as minor oxidized phase. After spark plasma sintering at 1000°C, the XRD of the sintered specimen includes the sharp double-peaks of solid solution γ phase and precipitate phase γ', which are very close to each other [13,14]. These results indicate that there was insufficient mechanical alloying effect in the ball milling for 5 h, but the γ and γ' phases in the sintered specimen could be formed by heat treatment at 1000°C.

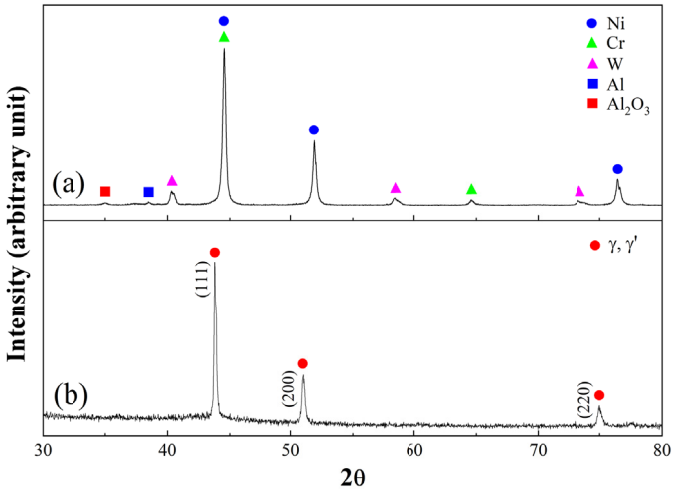

Fig. 4. XRD patterns of the Ni-based ODS superalloys at different stages of processing; (a) ball milling for 10 h and (b) spark plasma sintering at 1000°C

Fig. 5 shows the microstructures for Ni-based ODS superalloys that sintered using different PCAs. In the specimen ball-milled using stearic acid (Fig. 5a), inhomogeneous and large phases were observed. Conversely, Ni-based superalloy using ethanol (Fig. 5b) exhibited a homogeneous microstructure with fine precipitates at the grain boundaries. Considering that more uniform distribution of main elements was observed in the powder mixture ball-milled with ethanol, as shown in Fig. 3, these microstructural characteristics can be explained by the PCA effect including residual carbon content. These results indicated that the PCA strongly influenced the final microstructure of the sintered composites, and thus the Ni-based ODS superalloys with sound microstructure can be fabricated by using ball-milled powders with ethanol, more effectively.

4. Conclusions

The Ni-based ODS superalloys were fabricated from high-energy ball milling and spark plasma sintering process. Starting elemental powders were initially mixed in a tubular mixer and ball-milled with different PCA types of stearic acid and ethanol. The ball-milled powders for 5 h with ethanol showed low carbon content and more uniform distribution of main elements compared with the powders prepared by stearic acid. XRD analysis for the sintered specimen showed the peaks of solid solution γ and precipitate γ' phases. SEM observation revealed that the sintered alloy prepared by stearic acid exhibited inhomogeneous microstructure with large phases, while that prepared by ethanol showed a homogeneous microstructure with fine precipitates at the grain boundaries. Microstructure evolution of powder and sintered alloy has been discussed on the basis of mechanical alloying mechanism of powder mixtures with the different melting point and molecule size of PCA.

Acknowledgments

This work was supported by the Technology Innovation Program (10048158, Development of 980°C grade superalloys strengthened by multi-component nano-oxides for commercialization of core materials in the field of the defense industry) funded by the Ministry of Trade, Industry and Energy (MI, Korea).

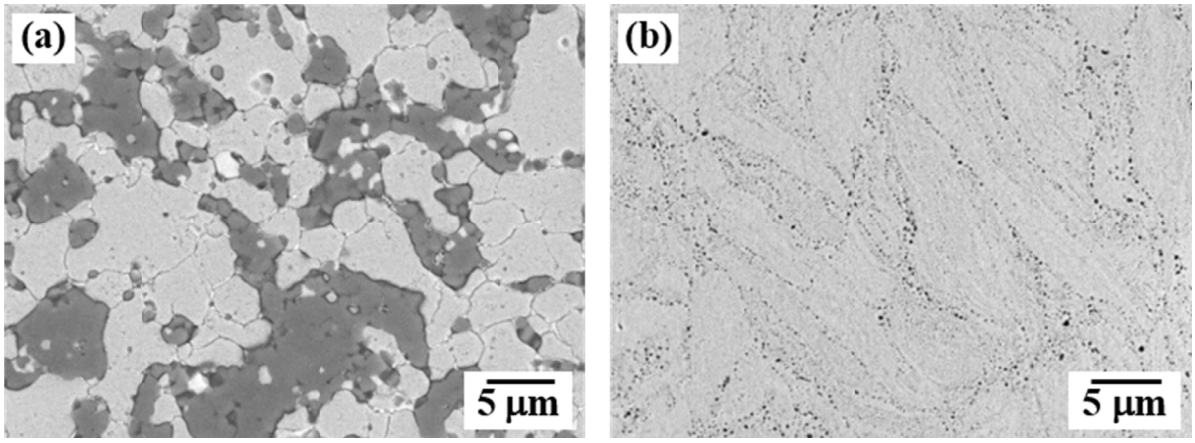

Fig. 5. SEM images (BSE mode) of the microstructure after spark plasma sintering of ball-milled powders with (a) stearic acid and (b) ethanol

REFERENCES

[1] J.S. Benjamin, Metall. Trans. **1**, 2943 (1970).

[2] C.W. Park, J.M. Byun, J.K. Park, Y.D. Kim, J. Korean Powder Metall. Inst. **23**, 61 (2016).

[3] D. Häussler, B. Reppich, M. Bartsch, U. Messerschmidt, Mater. Sci. Eng. **A309-310**, 500 (2001).

[4] S.K. Kang, R.C. Benn, Metall. Trans. A **18**, 747 (1987).

[5] C. Suryanarayana, Prog. Mater. Sci. **46**, 1 (2001).

[6] P.S. Gilman, J.S. Benjamin, Ann. Rev. Mater. Sci. **13**, 279 (1983).

[7] L. Ma, B.S.-J. Kang, M.A. Alvin, C.C. Huang, KONA Powder Part. J. **31**, 146 (2014).

[8] L. Shaw, M. Zawrah, J. Villegas, H. Luo, D. Miracle, Metall. Mater. Trans. A **34**, 159 (2003).

[9] Y.D. Kim, J.Y. Chung, J. Kim, H. Jeon, Mater. Sci. Eng. **A291**, 17 (2000).

[10] C.L. Chen, Y.M. Dong, Mater. Sci. Eng. **A528**, 8374 (2011).

[11] T. Peng, I. Chang, Powder Techn. **266**, 7 (2014).

[12] W. Lee, S.I. Kwun, J. Alloys Compd. **240**, 193 (1996).

[13] S.M.S. Aghamiri, H.R. Shahverdi, S. Ukai, N. Oono, K. Taya, S. Miura, S. Hayashi, T. Okuda, Mater. Charact. **100**, 135 (2015).

[14] S.M.S. Aghamiri, H.R. Shahverdi, S. Ukai, N. Oono, M.N. Ahmadabadi, T. Okuda, Mater. Lett. **161**, 568 (2015).