Harmonic structure formation and deformation behavior in a ($\alpha + \gamma$) two phase stainless steel

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Abstract. In the present work the harmonic structure design has been successfully applied for achieving a combination of high strength and high ductility, simultaneously, in a two-phase steel. The compacts of two-phase stainless steels with harmonic structure were prepared by controlling mechanical milling (MM) of pre-alloyed stainless steel powders followed by spark plasma sintering. The controlled MM leads to the formation of severely deformed “shell” region, wherein the subsurface region in the immediate vicinity of the powder surface consists of a nanocrystalline structure followed by the inner region consisting of dislocation cell structure. These severely deformed regions form fine-grained network during subsequent sintering, resulting in Harmonic structure. This networked structure displayed high strength, high ductility, and better uniform plastic deformation as compared to the homogeneous fine/coarse grained structure. Such a unique combination of properties in the two-phase stainless steel powder compacts was found to be associated with the ability of the harmonic structure to evenly distribute the strain during plastic deformation.

1. Introduction

Microstructural refinement has been widely accepted as an attractive method to improve mechanical properties of structural materials. In particular, grain refinement of metals and alloys exhibited significant improvement in the yield and ultimate tensile strengths. However, the homogeneous fine-grained microstructures exhibit poor ductility at room temperature. Interestingly, it has been found that a microstructure with bimodal grain size distribution is capable of providing higher strength without sacrificing ductility \[1-4\]. Recently, a novel microstructural design, called "Harmonic Structure Design", was proposed which resulted in a combination of high strength and high ductility at the same time \[5-10\]. A schematic representation of the harmonic structure design is shown in Figure 1. The harmonic structure design is essentially a bimodal microstructure consisting of a three-dimensional continuous network of fine-grained regions, called “shell”, which encircles the uniformly distributed coarse grained regions, termed as “Core”.

The harmonic structure design has been immensely successfully in achieving a combination of high strength and high ductility in materials with single phase microstructures \[7-10\]. However, the suitability and effectiveness of this novel microstructure design to a two-phase material has not been investigated so far. SUS329J1 is a typical two phase stainless steel, which is widely used as a piping material for seawater plants and marine structural material due to its excellent corrosion resistance \[11-13\]. Therefore, the present study deals with the
preparation, microstructural characteristics, and mechanical properties of harmonic structure SUS329J1 two-phase stainless steel. In the present work, compacts of two-phase stainless steels with harmonic structure were prepared by controlled mechanical milling of prealloyed stainless steel powders followed by spark plasma sintering of the milled powders. Spark plasma sintering is the method of sintering by electrical resistance heating and its most distinctive feature is a rapid sintering compared to other sintering methods. The microstructural characteristics and mechanical properties of the two phase stainless steel powder compacts, thus prepared, are presented and discussed.

2. Experimental Procedure
The SUS329J1 (Cr:25, Ni:4.5, Mo:2, Fe: Bal. (Mass%)) powder, used in the present work, was produced by Plasma Rotating Electrode Process (PREP). The as received pre-alloyed powder was in a fully-ferritic state, and had an average particle size approximately 280µm. The powder was mechanically milled (MM) for 90ks at ambient temperature under inert gas (Ar) atmosphere in a Fritsch P-5 planetary ball mill using SUS 304L vials and balls. The milling was carried out at 200rpm with ball-to-powder ratio of 1.8. Subsequently, the mechanically milled powder was sintered by Spark Plasma Sintering (SPS) process at 1123 K for 1.8ks under an applied pressure of 50MPa in vacuum. The microstructural characteristics of the MM powder and the sintered compacts were studied by Scanning Electron Microscope (SEM) and Transmission electron Microscope (TEM). Microstructural analysis was carried out by X-Ray Diffraction (XRD) and Electron Back Scatter Diffraction (EBSD) techniques. Mechanical properties were evaluated by Vickers Hardness tests and tensile tests. The tensile tests were carried out using a Shimadzu AGS-10kND tensile testing machine at an initial strain rate of 5.6x10^{-4} s^{-1}. Tensile test were performed on sub-size specimens with following gauge dimensions: 3 mm length, 1 mm width, and 1 mm thickness.

3. Results and Discussion
Figure 2 shows SEM images of SUS329J1 powders exhibiting the morphology and cross-section of the initial and 90ks MM powder particles. The initial PREP powders were spherical.

![Figure 2. Micrographs of initial and MMed SUS329J1 powders.](image-url)
with a smooth surface (Fig. 2a). On the other hand, the milled powder particles appeared as slightly deformed-spheres with rough surfaces due to the presence of depressions caused by the impact of balls during controlled mechanical milling (Fig. 2b). Clearly, the MM did not lead to either fragmentation or agglomeration of the powder particles. Figure 2 (c) and 2 (d) shows the cross-section of the initial and MM powder, respectively. It can be noticed that the initial powder particles had a typical coarse dendritic structure (Fig. 2c) whereas the milled powders consist of a deformed dendritic structure near the surface of the powder particles while the coarse dendritic microstructure is retained in the inner regions (Fig. 2d). These observations indicate that the plastic deformation during controlled milling was limited only to the areas near the surface of the milled powders. Figure 3 shows the a micrograph depicting the microstructure of the sub-surface region of the milled powder at higher magnifications. It evident that the deformed powders consisted of a thin layer, of the order of 15 µm thicknesses, of deformed dendritic structure near the surface of the illed powder. Also, the outer layer had a relatively higher hardness as compared to that of the inner region of the particle, which is an indication of the accumulation of defects due to the plastic deformation. However, it is envisaged that the degree of plastic deformation in the layer will not be uniform and the density of accumulated dislocations will have a gradient from the surface of the powders towards the inner region, i.e. dislocation density in the deformed layer will decrease from the surface towards the inner regions. Furthermore, the milled powder also exhibits the presence of a single phase microstructure.

Figure 4 shows the microstructure of the uppermost layer region of milled powder. It can be observed that in the immediate vicinity of the milled powder surface, i.e. at a distance ~ 1.5µm from the surface, the deformed layer consisted of equi-axed nano crystalline structure with average grain size of ~29 nm. The selected area diffraction pattern of the corresponding area exhibited Debye rings of α phase, indicating that these nano-sized crystallites are randomly distributed and separated by high angle boundaries. On the other hand, nanocrystalline structure could not be obtained in the region located at a little further away from the surface, at approximate 3µm from the surface, and cell structure existed. It appears that such a microstructural evolution occurred due to the existence of the gradient of the degree of deformation in the milled powder, i.e. strain accumulation and degree of plastic deformation decreases gradually from the surface towards the center of the milled powder. As the results, nano grains are generated in the uppermost layer region by mechanical milling.
Figure 5 shows typical EBSD grain size maps of the sintered MM powder compacts. It can be seen that the sintered compacts consisted of harmonic structure, i.e. a coarse grained region ("Core") surrounded by a continuously connected network of fine-grained regions ("Shell"). The core region is made up of a majority of grains with size more than 5 µm whereas the shell region had grains with size less than 5 µm. Figure 5 (b) shows an enlarged view of the area depicted as the Shell region in Figure 5 (a). It can be seen that the shell region consists of fine-sized equi-axis grains. It is also interesting to note that the Shell region itself consists of a grain size gradient, i.e. the middle of the Shell region consists of primarily sub-micron sized grains (represented as blue color) whereas “outer-shell” regions consists of grains of size 1-5 µm.

Figure 6 shows a typical TEM image of the shell region. It can be seen that the sub-micron grain structure exists in the center of the field, marked as the Mid-Shell, and micron size grain structure is located on both sides of Mid-Shell, which is termed as Outer-Shell. The average grain size of the mid-shell region and outer-shell region was 520 nm and 1.1 µm, respectively. Therefore, it is clear that the Shell region has a bilayer structure. This feature has been observed only in two phase materials, and is not typically observed in single phase materials. Therefore, it is considered a distinctive structure in harmonic structure two phase materials.
Figure 7 shows a TEM image depicting the microstructure and grain misorientations in Mid-Sell region. It is clearly evident that the Mid-Shell region consists of submicron-sized equi-axis grains separated by high-angle α grain boundaries. It appears that such a microstructural evolution occurred due to the existence of the nano-grains in the uppermost layer of the milled powder as the Mid-Shell region is formed by the extreme uppermost deformed layer present on the milled powders. Since the surface of the powder particles undergoes highest degree of plastic deformation, it consists of large amounts of accumulated dislocations and pre-existing nano grains. It appears that the γ precipitation at the HABs occurred in the very early stage of sintering, resulting in an ultra-fine (α + γ) microduplex structure.

Figure 8 shows the EBSD grain boundary angle map. White lines indicate low-angle boundaries (5-15 degree) and these are located and concentrated in the Outer-Shell region. The existence of low angle boundaries suggests that the Outer-Shell is the recovered structure, dominated by the low-angle grain boundaries. The inner part of the deformed layer on the milled powder consisted of very small amounts of accumulated defects due to decreasing effect of plastic deformation with increasing distance from the surface. Therefore, an exposure to an elevated temperature during spark plasma sintering leads to the grain growth of pre-existing nano-grains of the severely deformed areas whereas recovery occurs in the areas with relatively small degree of deformation. Furthermore, it appears that the γ phase precipitation suppress the recovery and recrystallization of the α matrix in the outer-shell region.

Figure 9 shows the tensile test results of the specimens prepared from conventional bulk duplex steel, compacts prepared from the as-received PREP powders, and compacts with harmonic structure. It can be clearly observed that the compacts prepared from initial powder had yield strength and ultimate tensile strength comparable to those of the specimen prepared from conventionally produced duplex steel. Moreover, it can also be noticed that the ductility of the initial powder compacts was inferior to that of the conventional steel. On the other hand, the harmonic structure compacts exhibited much higher yield and ultimate tensile strengths as compared to those of the conventional SUS329J1 steels. Moreover, the harmonic structured compacts also exhibited elongation to fracture comparable to that of the conventional material. Furthermore, it would also be worth emphasizing that the harmonic structured duplex steel compacts exhibited uniform elongation, of the order of 20%, in spite of significant strengthening. Therefore, it appears that the harmonic structure design is very
effective in achieving both high strength and good ductility in two-phase materials, such as \((\alpha + \gamma)\) duplex steels, also.

\[ \text{Figure 9 Tensile test results} \]

4. Conclusions

1. The harmonic structured two-phase stainless steel (SUS329J1) compacts were prepared by controlled milling and spark plasma sintering of PREP pre-alloyed powders. The controlled mechanical milling results in a thin deformation layer with nanocrystalline structure on the surface of the powders.

2. The harmonic structure compacts consisted of Shell and Core regions with \((\alpha + \gamma)\) two phase structure. Furthermore, the shell region consisted of two grain size regimes namely, (i) Mid-Shell region with submicron-sized grains, and (ii) Outer-Shell with 1-5 µm size grains. Such a microstructural evolution was attributed to the existence of the gradient in the degree of deformation in the deformed outer layer of the milled powder.

3. The sintered steel compacts with harmonic structure exhibited significantly improved mechanical properties as compared to the conventional bulk steel and initial powder compacts.

4. It has been demonstrated that the harmonic structure design can be effectively used to achieve a combination of high strength and good ductility at the same time in the two-phase materials also.

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