



SURFACE NANOCRYSTALLIZATION OF IRON INDUCED BY ULTRASONIC SHOT PEENING

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Abstract—A nanocrystalline surface layer was fabricated on a pure Fe plate by using an ultrasonic shot peening (USSP) technique. The USSP induced surface nanocrystallization products were characterized by means of X-ray diffraction (XRD) and transmission electron microscope (TEM). Experimental evidences showed that after USSP treatments, the initial coarse-grained structure in the surface layer was refined into equiaxed ultrafine grains (about 10nm) with random crystallographic orientations. The grain refinement mechanism during USSP treatments is discussed. ©1999 Acta Metallurgica Inc.

Introduction

Nanocrystalline (nc) materials, that are structurally characterized by ultrafine grains (typically less than 100 nm in at least one dimension) and a large volume fraction of defective grain boundaries, have attracted increasing scientific interests in recent years. Numerous experimental results have indicated that nc materials possess properties and behaviors that are fundamentally different from, and often superior to, those of their conventional coarse-grained polycrystalline counterparts (1–4). Nanocrystallization of a coarse-grained polycrystalline material provides a new approach to the improvement of properties without changing its chemical composition. Therefore extensive and intensive research activities in the area of nc materials have been carried out that are driven by both a need for basic understanding of the nature of nanostructures and the potential technological applications of nc materials with their unique properties.

Currently, most research on nc materials deals with 3-dimensional bulk metals and ceramics with regard to their synthesis and processing, structural characteristics and properties. Various kinds of synthesis techniques have been developed for producing 3-D bulk nc samples, such as inert gas condensation and consolidation (IGC) (1), severe plastic deformation (SPD) (5), crystallization of amorphous solids (3) and electrodeposition (6). With these methods, different 3-D nc materials have been successfully prepared that are essential to understand the intrinsic structure and properties of polycrystalline materials with ultrafine grains. Up to the present, at least two major difficulties still exist in synthesizing 3-D nc materials using current techniques: (1) It is difficult to synthesize “ideal” 3-D nc samples, i.e., that are porosity-free, contamination-free and of sufficient size for conventional

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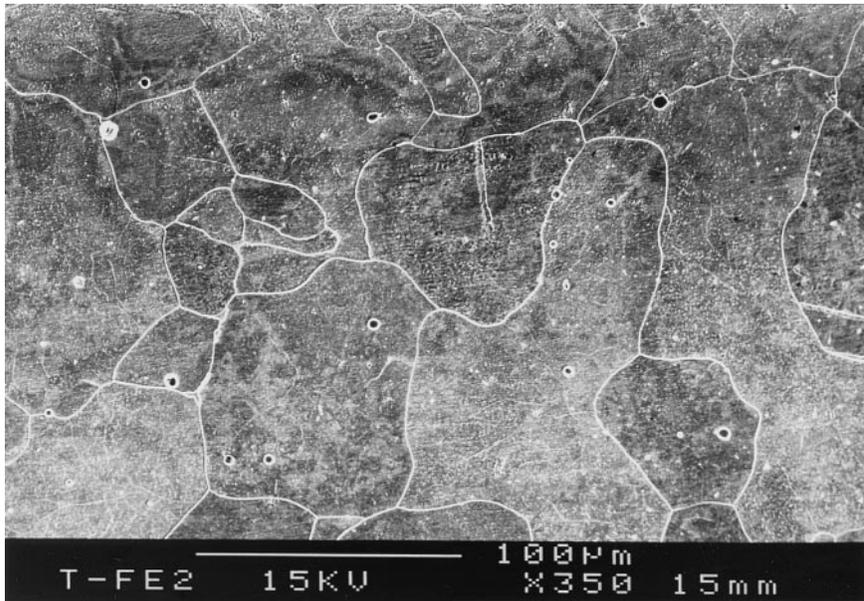


Figure 1. SEM micrograph of the coarse-grained Fe specimen (annealed state) before USSP treatments.

property measurements for revealing the intrinsic nanometer-grain-size effect on the structure-property relationship without any external influencing factors. (2) Most of the current techniques for synthesizing nc materials are difficult to transfer into industrial-scale applications, due to their limitations in cost, sample size and types of materials.

It is known that material failures occur on the surface, in most cases. Most material failures, including fatigue fracture, fretting fatigue, wear and corrosion, are very sensitive to the structure and properties of the material surface. Optimization of the surface structure and properties may effectively improve the global behavior of the material. With the increasing evidence for nc materials' unique properties, we proposed to achieve surface modification by generation of a nanocrystalline surface layer so that the overall properties of the material might be significantly improved (7). This kind of surface modification, called surface nanocrystallization (SNC), will provide a new approach that makes it possible to meet specific structure/property requirements on the surface of the material.

In this paper, we will present a new technique for forming SNC on metallic materials by using pure iron as an example. The method is based on mechanically-induced grain refinement using an ultrasonic shot peening (USSP) method (8). A layer of nc structure was successfully obtained on pure Fe samples, which was characterized by x-ray diffraction (XRD) and transmission electron microscopy (TEM). The grain refinement mechanism during the mechanical processing is discussed.

Experiment

An industrial-pure iron plate was subjected to USSP in order to achieve a SNC structure. The plate was $10 \times 60 \times 60 \text{ mm}^3$ in size, of which the surface was first polished with silicon carbide papers and then vacuum-annealed at 923 K for 120 min. The grain size of the annealed Fe plate is between 20 ~ 150 μm , as seen in Figure 1.

Figure 2 shows a schematic illustration of the USSP treatment set-up. The stainless steel shots, with a diameter of 3 mm, were placed in a reflecting chamber (including an ultrasonic concentrator) that was

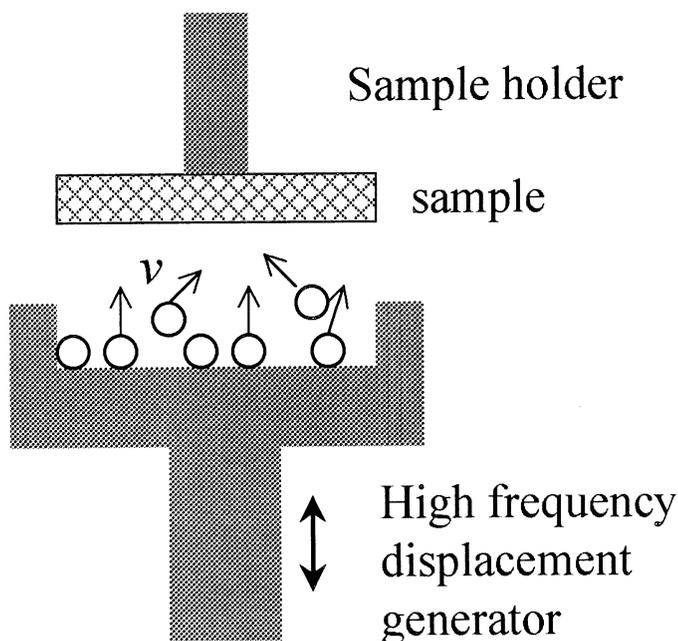


Figure 2. Schematic illustration of the ultrasonic shot peening treatment set-up.

vibrated by an ultrasonic generator, after which the shots were resonated. Because of the high frequency of the system (20 kHz), the entire surface of the sample to be treated was peened with a high number of shots in a short period of time. The shot-peening intensity induced by this technique is similar to that of a conventional shot-peening method. In the present work, the surface of the Fe plate was treated by using USSP methods at room temperature for 50, 150, 450 and 1250 s, respectively.

The X-ray diffraction analysis of the Fe surface layer was carried out on a Rigaku D/max 2400 X-ray diffractometer (12 kW), with Cu $K\alpha$ radiation, in the step-scanning mode. X-ray wavelengths $\lambda_{K\alpha1} = 1.54056 \text{ \AA}$ and $\lambda_{K\alpha2} = 1.54439 \text{ \AA}$ were selected, using a graphite slit section. The calibration of the instrumental line broadening was performed by using a standard SiO_2 sample. The experimental temperature was $293 \pm 1 \text{ K}$. The average grain size and the mean microstrain were calculated in terms of the diffraction line broadening of six single Bragg reflection peaks of bcc Fe (110), (200), (211), (220), (310), and (222) by using the methods which were described in (9). The lattice parameters were calculated from the intensity peak top positions for the Fe samples. The peak positions were calibrated by the external standard method, using a pure Si polycrystal in order to minimize errors in the system. $\text{Sin}^2 2\theta$ was selected as the error function. All six single peaks were used in the calculations.

The TEM experiments were performed on a Philips EM 420 transmission electron microscope with an operating voltage of 120 kV. Thin foil samples for TEM observations were cut from the Fe surface by using an electro-spark discharge machine and thinned by using ion milling in a vacuum at low temperature.

Results and Discussion

The XRD patterns of the Fe samples before and after USSP treatments are shown in Fig. 3. It was observed that after USSP there were an evident broadening of the Bragg reflections and a shift in the centroid position of diffraction peaks relative to the coarse-grained sample. This might be attributed to

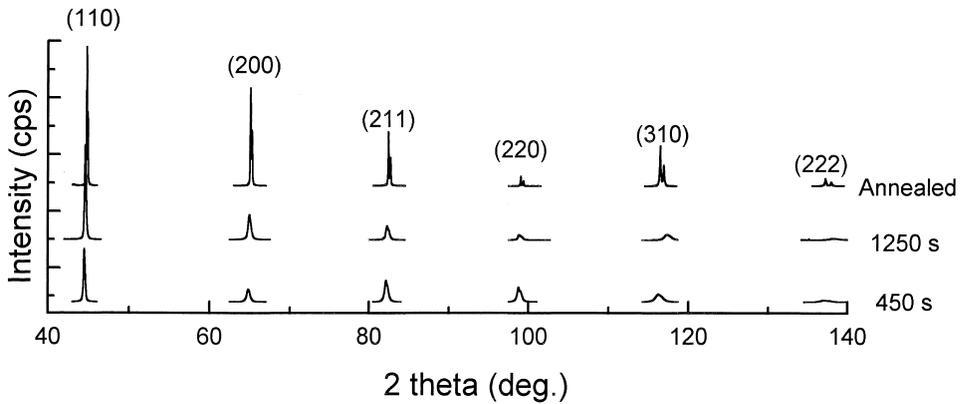


Figure 3. X-ray patterns of the Fe samples after USSP for various periods of time (as indicated) and the coarse-grained Fe specimen after annealing.

grain refinement and microstrain development, and/or micro-distortions of the crystalline lattice. It was noted that the XRD patterns of the Fe after USSP treatments were similar in many ways to those of the nc materials obtained by IGC (10) and the ultrafine-grained materials obtained by SPD (11). The structural parameters obtained by the calculations of XRD data are listed in Table 1. The average grain size of the surface layer after USSP treatments was found to be about 30 nm when the treatment duration was between 50–450 s. With a longer treatment time (1250 s), the average grain size was increased to about 37 nm. The microstrain derived from XRD measurements was found to be about 0.03 ~ 0.08%, which was much smaller than that of the nc materials synthesized by means of the consolidation of ultrafine powders and SPD methods (up to 3%). The lattice parameter of the nc layer differs from the equilibrium lattice constant ($a_0 = 2.8663 \text{ \AA}$). This might originate from lattice distortions in nc phases, analogous to the observation in other nc materials (12).

Figures 4 and 5 show the TEM micrographs and the statistic grain size distribution for the samples after USSP treatments for 450 and 1250 s, respectively. In both the bright and the dark field images, one can see uniform ultrafine grains that are mostly equiaxed. These nc grains possess random crystallographic orientations, as indicated in the selected area electron diffraction (SAED) patterns. The histogram of grain size distribution obtained from the dark field image of the nc Fe surface layer was characterized by a normal logarithmic distribution with a narrow size distribution, as shown in Fig. 4d and Fig. 5c, respectively. The mean grain size of the surface was approximately 10 nm and 16 nm in the samples peened for 450 and 1250 s, respectively. The grain size determined from TEM observations was evidently smaller than that obtained from the XRD calculations. This might be due to the fact that XRD results averaged the structure information of a surface layer of about $10 \mu\text{m}$ thick, while the TEM

TABLE 1
XRD Measurement Results of the Average Grain Size, Mean Microstrain and Lattice Parameter in the Fe Samples after USSP Treatments for Different Periods of Time

Peening Time (s)	D (nm)	$\langle \varepsilon^2 \rangle^{1/2}$ (%)	a (\AA)
50	25 ± 3	0.051 ± 0.004	2.8686 ± 0.0002
150	30 ± 3	0.050 ± 0.003	2.8699 ± 0.0006
450	28 ± 4	0.035 ± 0.004	2.8705 ± 0.0007
1250	37 ± 3	0.083 ± 0.002	2.8682 ± 0.0003

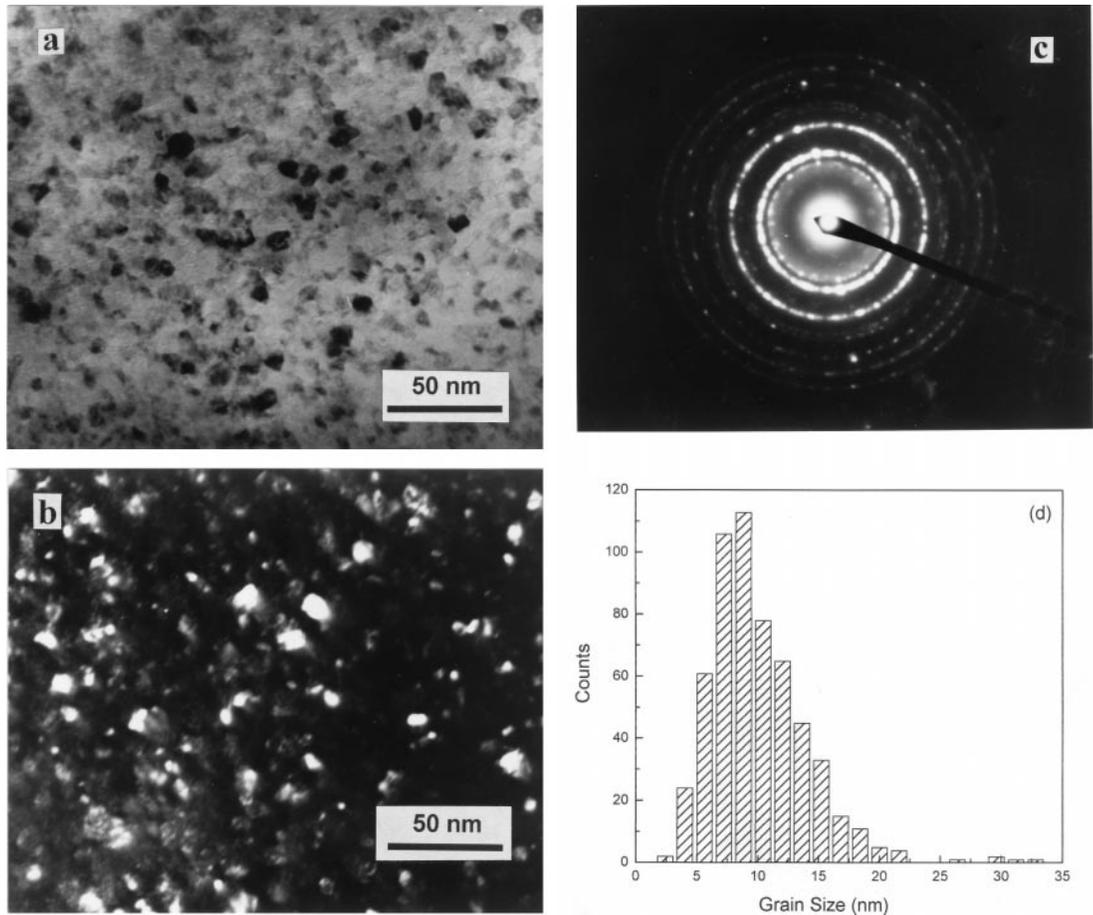


Figure 4. Bright-field (a) and dark-field (b) TEM images, selected area electron diffraction (c) and statistic grain size distribution derived from TEM observations (d) of the surface layer of Fe specimen after USSP for 450 s.

sample was a very thin film (less than $1 \mu\text{m}$ thick) at the sample surface. There was a grain size change which progressed with the distance from the peened surface.

TEM observations indicated that the grain size evidently increased with an increment of the distance from the peened surface. Figure 6 shows the microstructure in a layer at about $10 \mu\text{m}$ deep from the surface peened for 450 s. The grains (about $50 \sim 100 \text{ nm}$) are much larger than those in Fig. 4. The grain boundaries between these large crystallites are not as distinct as those between small grains at the peened surface. These results indicated that a nc layer at least $10 \mu\text{m}$ thick had been formed on the Fe surface after USSP treatments for 450 s.

With an extension of the USSP time to 1250 s, the average grain size at the peened surface was increased, as could be seen from the XRD and TEM experiments. Although the intrinsic reason for the grain size increment is not clear yet, the temperature raise during shot peening for a longer time was observed which might be responsible for elevating the stable grain size with which a balance between dislocation generation and its recovery is established.

Formation of nc structure in the surface layer of the Fe during USSP treatments might be understood as analogous to the nanocrystallization process during the severe plastic deformation of metals and alloys (7). The USSP process provides repeated multidirectional mechanical loads at high speeds onto

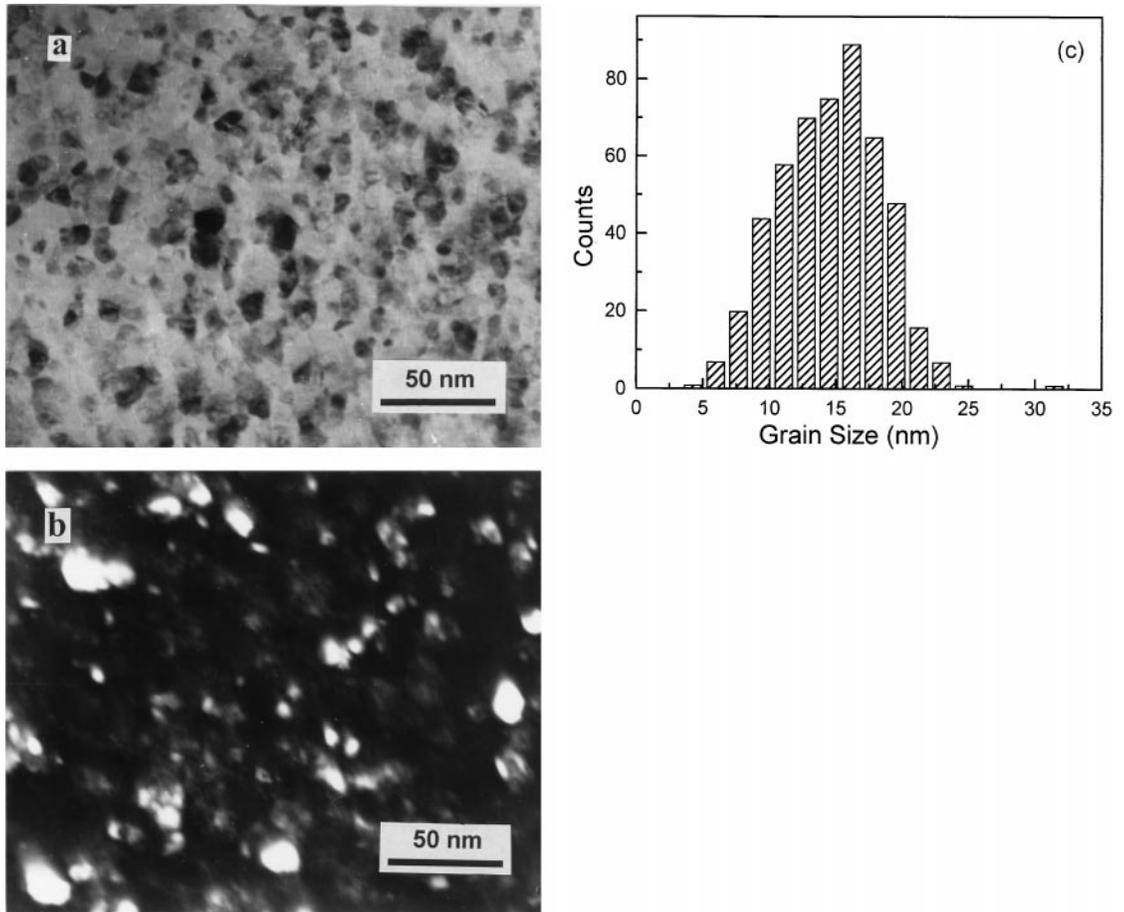


Figure 5. Bright-field (a) and dark-field (b) TEM images, statistic grain-size distribution derived from TEM observations (c) of the surface layer of Fe specimen after USSP for 1250 s.

the material surface. The peening loads will generate dislocations and eventually result in plastic deformation in the surface layer of the material. The plastic deformation might be localized in multiple shear bands in the surface layer, due to the differences in loading direction of the shots. Multiple shear bands have been observed in the surface layer of a stainless steel (fcc structure) after USSP treatments (13). The shear bands consist of high-density dislocation arrays. Repeated shot peening creates more dislocations which will be annihilated or recombined (rearranged) to form small angle grain boundaries separating individual grains, as proposed by Fecht in analyzing the grain refinement of metals during ball milling (14). Further USSP treatments may induce changes in the orientation of the grains with respect to their neighboring grains, forming crystallites with completely random orientation. Such a grain refinement process may result in the formation of ultrafine (nm-sized) grains, depending on the load intensity and strain rate. With an increase in the peening time, a nc layer will be developed and its thickness increases.

The surface nanocrystallization mechanism as proposed above needs to be verified with more experimental evidence. Direct high-resolution TEM observations of the microstructure evolution during USSP processing will be helpful in obtaining a better understanding of the surface nanocrystallization processes, that are currently being developed.

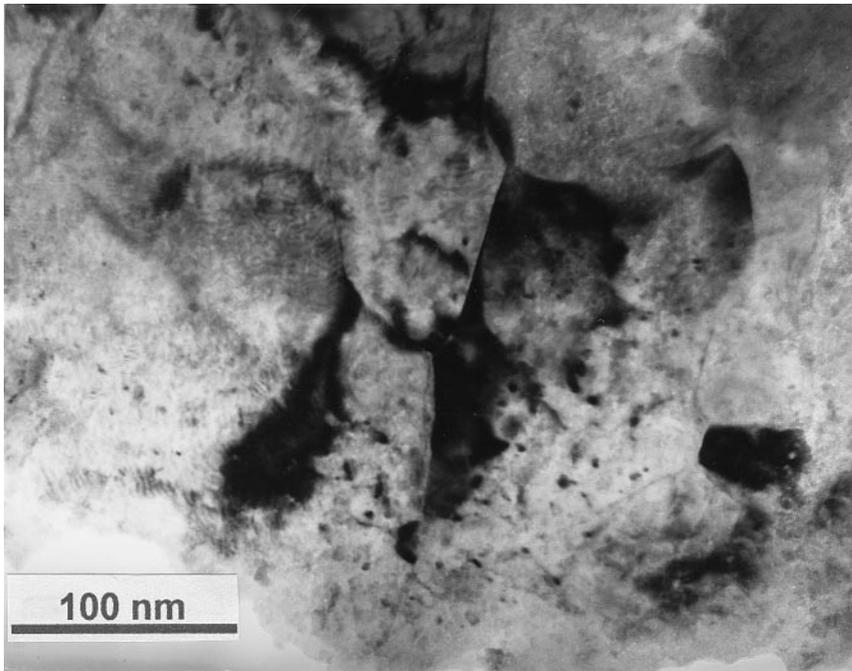


Figure 6. A bright-field image of the layer at a distance of about 10 μm from the peened surface of the Fe sample peened for 450 s.

Conclusions

Nanocrystalline structure was successfully obtained on the surface layer of a pure Fe plate by means of USSP treatments. The average grain size of the surface layer after USSP treatments was as small as 10 nm. This work demonstrates that the USSP treatment provides an effective approach for forming a nanostructure layer on the surface of metallic materials that may find applications in improving the mechanical and corrosion-resistant properties.

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