*Journal of the Korean Society
for Nondestructive Testing
Vol. 28, No. 3 (2008. 6)*

Experimental Characterization of Cyclic Deformation in Copper Using Ultrasonic Nonlinearity

C. S. Kim*, Ik-Keun Park**†, Kyung-Young Jhang*** and Nohyu Kim****

Abstract We have experimentally investigated the cyclic deformation in copper using ultrasonic nonlinearity. The observation and characterization of dislocation substructure have been conducted using transmission electron microscope and electron backscattered diffraction technique. The ultrasonic nonlinearity (β/β_0) was measured by the harmonic generation technique after various fatigue cycles. The microstructural effect on the nonlinearity was discussed regarding the extent of dislocation substructures evolved from low cycle fatigue. The ultrasonic nonlinearity of copper monotonically increased with the fatigue cycles due to the evolution of dislocation cell substructures.

Keywords: Ultrasonic Nonlinearity, Dislocation, Harmonic Generation Technique, Fatigue

1. Introduction

Structural materials subjected to cyclic loading are well known to be closely related to the fatigue crack initiation and propagation to the failure. Before crack initiation state, materials evolved change their mechanical properties at the beginning of the fatigue process. Such changes in mechanical properties may be closely related with dislocation density and configuration. Dislocations build up that lead to the formation of the persistent slip band (PSB). The slip on the most active slip system should be directed at the intersection of the grain boundary with the specimen surface. These slip bands act as stress concentrators for the origination of microcracks, which can grow under cyclic loading and lead to catastrophic failure of materials (McEvily, 1979).

Therefore, structural components that are subjected to fatigue stresses should regularly undergo safety evaluation. Moreover, a fundamental understanding of the microstructural changes that occur during the plastic deformation process, such as the dislocation density and substructure, is crucial for the nondestructive evaluation of these materials.

The ultrasonic nonlinearity technique has been found to strongly sensitive with microstructures (Cantrell and Yost, 2000; Meyendorf et al., 2004; Kim et al., 2005; Kim and Park, 2008). Nonlinear elasticity of solid relates to forces acting between atoms in crystals, which are the interatomic potentials characterized by the well depth and equilibrium interatomic separation. Interatomic potentials in real crystals are not harmonic, and the anharmonicity can be treated by higher order elasticity. The potentials are determined by measurement of the sublimation energy of the crystal and of its lattice spacing. When a

---

Received: May 6, 2008, Revised: June 16, 2008, Accepted: June 19, 2008. * Engineering Science and Mechanics, Pennsylvania State University, 212 EES Building, State College, PA 16802, USA, ** Department of Mechanical Engineering, Seoul National University of Technology, Gongneungdong Seoul 139-743, Korea, *** School of Mechanical Engineering, Hanyang University, Haengdangdong Seoul 133-791, Korea, **** School of Mechatronics Engineering, Korea University of Technique & Education, Cheonan-si 330-708, Korea † Corresponding Author: ikpark@snut.ac.kr

sinusoidal ultrasonic wave of a given frequency and of sufficient amplitude is introduced into an anharmonic solid, the fundamental wave may be distorted as it propagates, so that the second and higher harmonics of the fundamental frequency will be generated. Measurement of the amplitude of the second harmonic as a function of the amplitude of the fundamental frequency has been carried out by Breazeal and Tompson for the purpose of understanding the material anharmonicity (Breazeale and Thompson, 1963). Breazeale and Ford studied the nonlinear behavior of ultrasonic wave in copper single crystals which have been neutron irradiated through the use of finite amplitude distortion in order to observe the nonlinear properties of the crystal lattice alone (Breazeale and Ford, 1965). Hikata et al. investigated the dislocation contribution of the nonlinearity using the aluminum single crystal, and reported that the amplitude of second harmonic increased as a function of tensile stresses (Hikata et al., 1963). Cantrell presented an analytical model that the dependence of acoustic nonlinearity in wavy slip pure metal on the dislocation monopoles and dipoles (Cantrell, 2006). He reported that the acoustic nonlinearity increased with the fatigue cycles in martensitic 410C steel. Hurley et al. studied the nonlinear ultrasonic parameter in quenched martensite steels, which the mass percent of carbon varied from 0.1% to 0.4%. They reported that the nonlinear parameter increased monotonically with the dislocation density (Hurley et al., 1998).

In the previous reports, they have studied ultrasonic nonlinearity in the context of numerical and analytical modeling in order to understand the dislocation effects on the ultrasonic nonlinearity. Besides, they have applied to the carbon steels to the verification of their model without considering the precipitate effects on the nonlinearity. However, the second phase may be occur microstrain due to the mismatch between the matrix and precipitate phase, and hence can influence the distortion of ultrasonic waves. In contrast to these studies, we would like to investigate ultrasonic nonlinearity with a pure copper in order to eliminate the precipitate effects on the nonlinearity.

2. Acoustic Nonlinearity

The nonlinear effect closely relates to the higher order elastic constants in the elastic potential energy (strain energy), and the stress (σ) is expanded with differentiation of potential energy. If we assume that the attenuation can be neglected, then the equations of motion for longitudinal wave in solid that may be written as (Wallace, 1967):

$$\rho\frac{\partial^2 u}{\partial t^2}=\frac{\partial\sigma}{\partial x} \tag{1}$$

where u is the displacement in the x-direction, t is time and ρ is mass density of the solid.

Considering the one dimensional wave propagation of a pure longitudinal wave in isotropic solid, one can get as follows (Cantrell, 1994):

$$\frac{\partial^2 u}{\partial t^2}=C^2(1-\beta\frac{\partial u}{\partial x})\frac{\partial^2 u}{\partial x^2} \tag{2}$$

where $C=\sqrt{(E/\rho)}$ is the longitudinal wave velocity, E is Young's modulus and β is nonlinearity parameter.

An approximate solution of eqn. (2) is $u=u_0+u_1$ where u_0 is a solution with $\beta=0$, then the solution of eqn. (2) can be obtained by an iterative process (Melngailis et al., 1963):

$$u(x,t)= A_1\sin(kx-\omega t)+\frac{A_1^2k^2\beta x}{8}\cos 2(kx-\omega t) \tag{3}$$

where A_1 is particle displacement amplitude of fundamental wave, and for second harmonic wave A_2 is $(\beta A_1^2k^2x)/8$, k is the propagation constant $2\pi/\lambda$ where λ is the wavelength, $\omega=2\pi f$ where f is the frequency.

3. Experimental Details

3.1 Materials and Fatigue Test

The materials used in this study were 99.9% pure polycrystalline copper. All specimens were machined to have a gage length of 12 mm, and were annealed in an argon atmosphere for one hour, at 450℃. The final grain size, as determined using an average linear intercept method, were approximately 41 ㎛. The fatigue test specimens were carefully prepared by electropolishing in order to eliminate the surface damaged layer during the mechanical processing. The fatigue experiments were performed under constant strain amplitude at room temperature, using a servohydraulic fatigue testing machine. A triangular wave signal was used for the constant strain rate of 6×10^{-4}/s and the cycles to failure (N_f) was defined as the number of cycles at which the saturation stress decreased to 20%. A Manson-Coffin law (Hertzber, 1976) was curve fitted to measure the fatigue properties of copper.

$$\varepsilon_{ap} = \varepsilon_f'(2N_f)^c \tag{4}$$

where ε_{ap} is the plastic strain, ε_f' the fatigue ductility coefficient, N_f the cycles to failure and c the fatigue ductility exponent.

The monotonic and cyclic tests were also conduced in order to identify the material properties. The incremental step test (0% ~ 1%) was achieved with the same conditions of low cycle fatigue.

3.2 Acoustic Nonlinearity

When a sinusoidal ultrasonic wave of a given frequency and of sufficient amplitude is introduced into an anharmonic or nonlinear solid, the fundamental wave is distorted as it propagates, so that the higher harmonics of the fundamental frequency are generated. The piezoelectric *f-2f* method was used to measure the acoustic nonlinear parameter. The nonlinear parameter measurement system (RAM10000, RITEC Inc.) was primarily composed of a high power attenuator (RA-31), a high power 50 Ω termination (RL-50), and a high power 6 dB attenuator (RA-6). A longitudinal piezoelectric transducer with a nominal frequency of 10 MHz was used to generate the fundamental wave, and a wide band piezoelectric transducer with a 20 MHz was used in the receiving second higher harmonic wave. The waveform was digitally processed using power spectrum Fast Fourier Transformation, in order to obtain the displacement amplitudes, A_1 at the fundamental frequency and A_2 at the second harmonic frequency. The magnitude of the nonlinear parameter (β) was determined from the eqn. (3) indicates that the β may be obtained from measurements of the absolute amplitudes of the fundamental acoustic wave and second harmonic wave. Therefore, nonlinearity parameter is determined as follows:

$$\beta = \frac{8A_2}{A_1^2 k^2 x} \tag{5}$$

In this study, we measured the normalized ultrasonic nonlinearity parameter (β/β_0) in order to evaluate the extent of fatigue damage, where β is the nonlinearity parameter of fatigue damaged specimens and β_0 is the nonlinearity parameter of an as-annealed specimen. As for the ultrasonic nonlinearity test specimen, the copper materials were machined from a rectangular shape measuring 12 mm long, 10 mm wide and 10 mm thick.

3.3 Microstructural Evaluation

All cyclically deformed specimens were cut so the stress axis would be perpendicular to the plane of observation. All thin foils were prepared by electropolishing in a solution of 33% nitric acid and 67% methyl alcohol at a temperature of −25℃ and a voltage of 2 V, using a Struers

Tenupol-3 jet polisher. The foils were examined using a JEOL 1200EX transmission electron microscope. All TEM images were taken under bright field conditions.

For the electron backscatter diffraction (EBSD) observation, the surface of specimens were polished with 3 μm diamond paste, and with 0.5 μm alumina slurry, and then electropolished using a mixture of 700 ml phosphoric acid and 300 ml distilled water, which was cooled in dry iced water and stirred during polishing. The polishing conditions were 1.5-2 V for 5-10 minutes. A crystal orientation measurement was made with a TSL EBSD system, interfaced to a JEOL JSM-6500F, with a field emission electron gun (FEG system). This system can automatically obtain a crystal orientation map by scanning a rectangular area on the surface of the specimens tilted at 70° from the horizontal.

4. Results and Discussion

The strain hardening was observed to be most pronounced during the early stage of cycling, where after the strain hardening became saturated. Table 1 shows the monotonic and cyclic test results. The fatigue ductility coefficient (ε'_f) for copper is 0.35 and the fatigue ductility exponents (c) is -0.36 as evaluated using the least-squares fit of Manson-Coffin law.

Fig. 1 shows the ultrasonic nonlinearity of copper as a function of fatigue cycles. The nonlinearity parameter monotonically increased due to the fatigue driven plastic deformation. Generally, if an ultrasonic sinusoidal wave of a given frequency and of sufficient amplitude is introduced into an anharmonic solid, the fundamental wave may be distorted as it propagates, so that the second and higher harmonics of the fundamental frequency will be generated. Therefore, in this study, the fatigue driven plastic deformation is a dominant effect on the distortion of ultrasonic wave during its propagation through the specimens. In order to understand the microstructural features influenced on the distortion of ultrasonic wave, we note the microstructural evolution during fatigue damage, and then interpret the variation in the ultrasonic nonlinearity.

Fig. 2 shows the changes in the dislocation substructures of the copper after different numbers of fatigue cycles. The type of

Table 1 Comparison of monotonic and cyclic stress strain curves for incremental step test of copper at a strain rate of $6\times10\ ^{4}s\ ^{1}$

UTS	YS	c	e'f	Strength coefficient		Strain hardening	
				monotonic	cyclic	monotonic	cyclic
205	40	-0.36	0.35	305	334	0.38	0.12

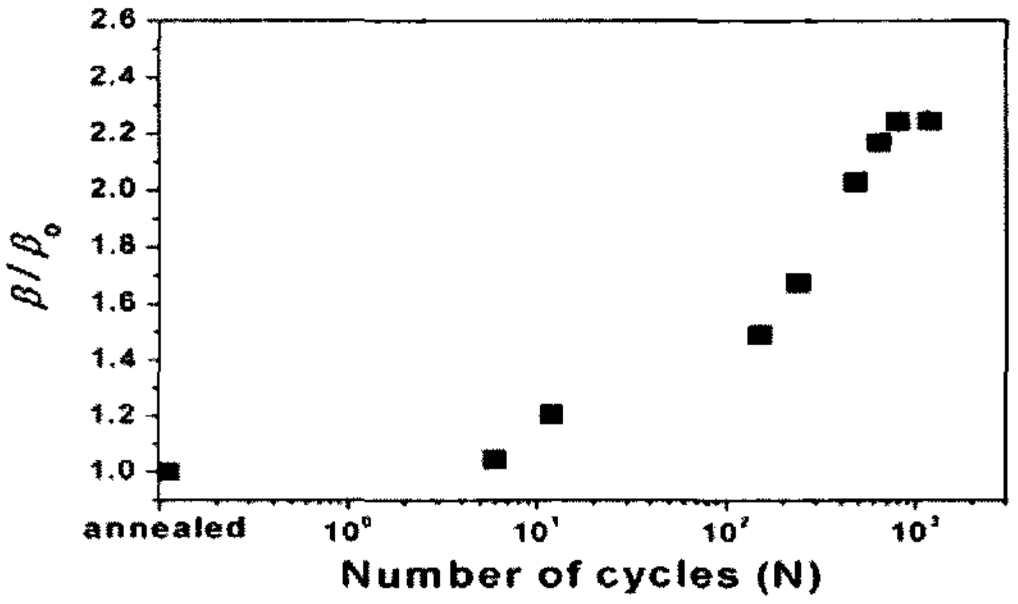

Fig. 1 Ultrasonic nonlinearity (β/β_0) of fatigue damaged copper as a function of number of cycles

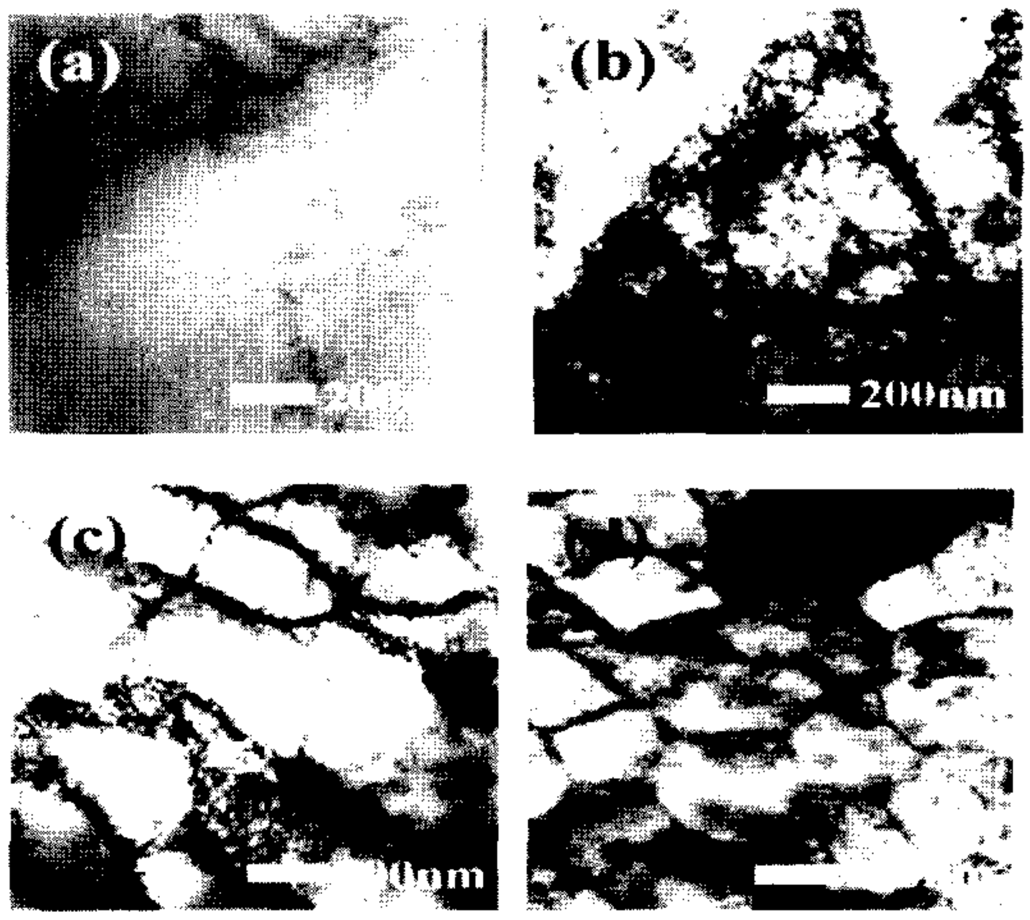

Fig. 2 TEM micrographs showing dislocation cell substructure of copper under the strain amplitude of 0.3%; (a) as annealed, (b) 240 cycles, (c) 640 cycles and (d) 1190 cycles

dislocation substructure found in the whole volume of the fcc specimens depends very strongly on the strain amplitude, the stacking fault energy, and, to a limited extent, the temperature (Madhoun, 2003). Copper is a wavy slip material, in which cross slip takes place easily as a result of the high stacking fault energy, and multiple slip leads to the decomposition of the persistent slip band into cellular bands, and then to a cell structure at a high strain amplitude (Feltner and Laird, 1967). In this study, the development of a dislocation cell substructure in copper was clearly observed. The results obtained correspond well to those previously reported by other researchers. The dislocation density on the cell structure cannot be reliably determined when using thin foils, because the dislocation density in the cell walls is too high, notwithstanding the slight decrease in cell size with increasing fatigue life fraction.

Fig. 3 represent image quality (IQ) map of copper under strain amplitude of 0.3%. The IQ describes the quality of an electron backscatter diffraction pattern, which is dependent on the material and its condition (Kim, 2007). The factor affecting the quality of diffraction patterns is the perfection of the crystal lattice in the diffraction volume. Thus, any distortions to the crystal lattice within the diffraction volume will produce lower quality diffraction patterns. As the fatigue life increased, the darker gray shades in the images continuously increased, indicating the stain distribution of copper. Fig. 4 shows the number fraction of the grain average misorientation (GAM) for copper as a function of fatigue cycles. The GAM is the algebraic average of the misorientation between all points and their nearest neighbor measurement points. The GAM will be low for structures with no geometrically required dislocations that accommodate lattice rotation (Trivedi, 2004). Deformation of a grain is dependent on the dislocation content, crystal lattice orientation, grain size and shape, second particle content and the orientations of neighboring grains. The crystal lattice orientation has a primary influence on the evolution of the dislocation substructure. Hence, the GAM was measured in the present investigation in order to study the effect of the crystal lattice orientation on the evolution of the dislocation substructure. As presented in Fig. 4, the GAM was clearly increased during fatigue deformation.

In general, the higher harmonic components were typically generated by the distortion of the ultrasonic wave during propagation in the

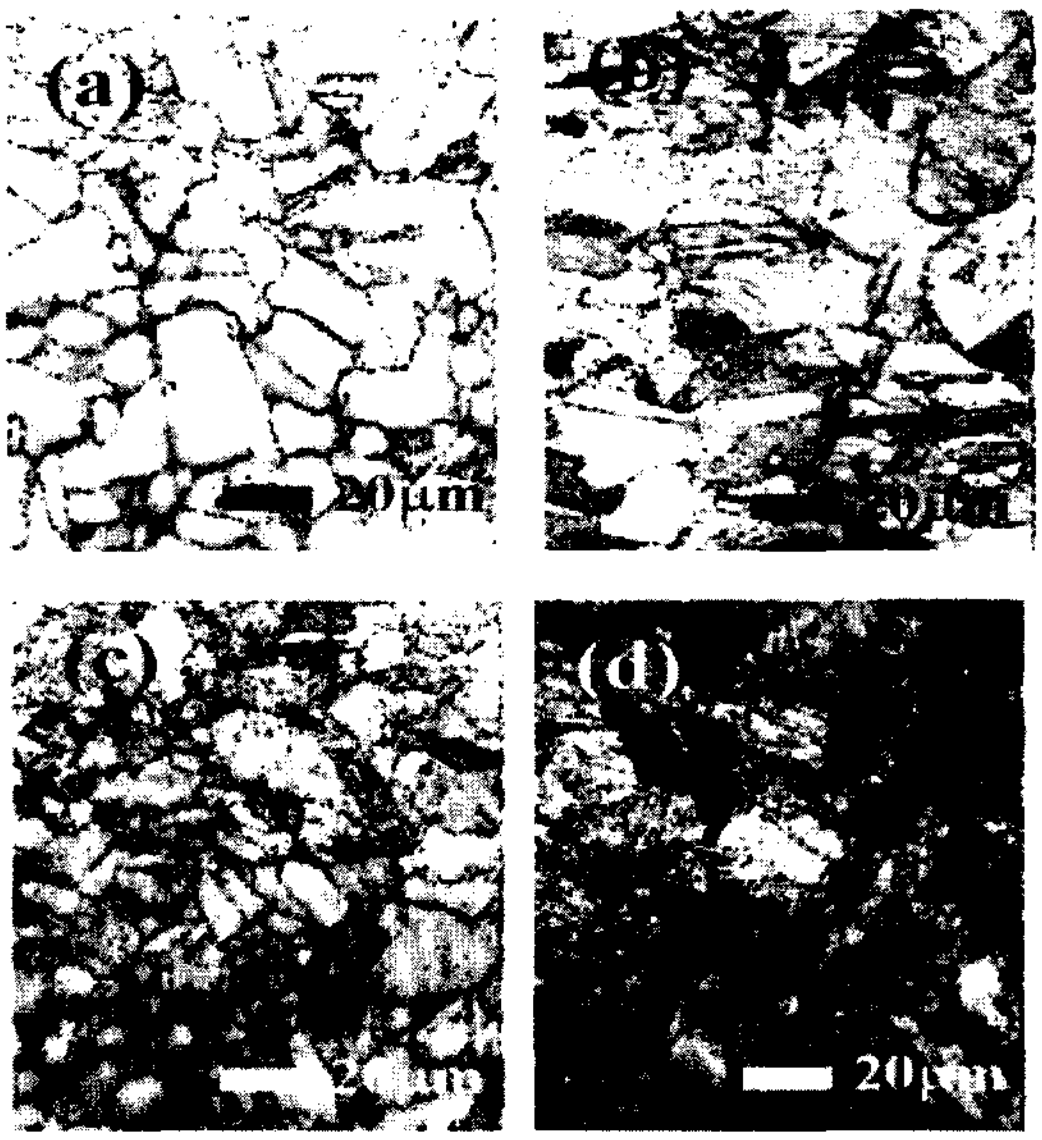

Fig. 3 Image quality map of copper under the strain amplitude of 0.3%; (a) as annealed, (b) 240 cycles, (c) 640 cycles and (d) 1190 cycles

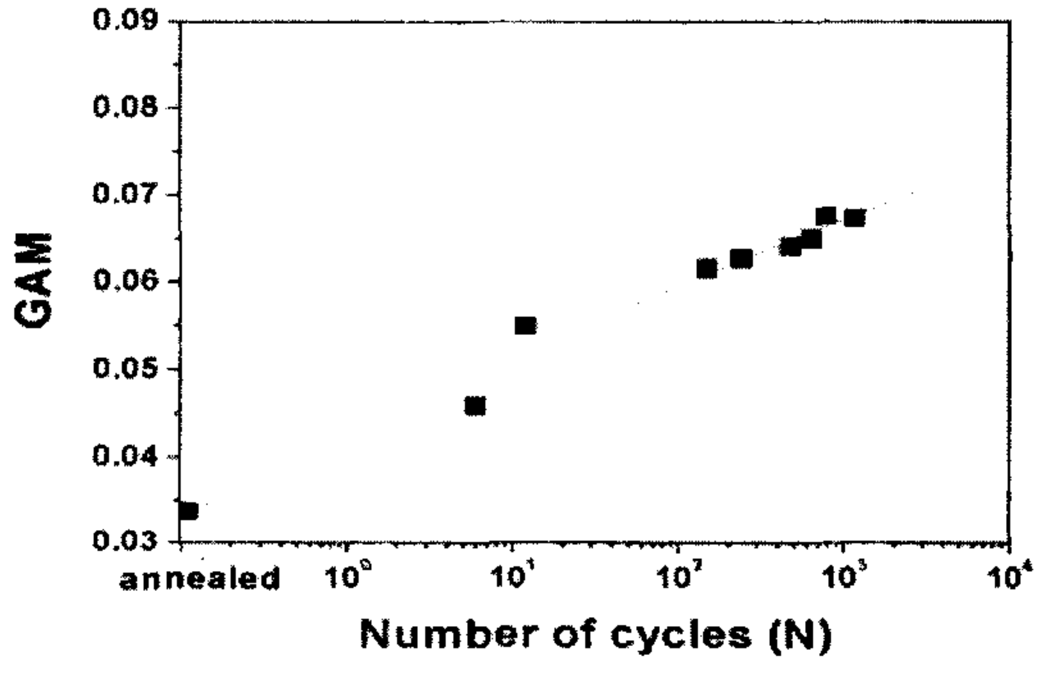

Fig. 4 Variation of grain average misorientation as a function of number of cycles

anharmonic solid, a phenomenon that is related with the lattice anharmonicity of crystals (such anharmonicity comes, for example, from phonon-phonon interaction, thermal vibrations, etc.). Another microstructural factor that can influence the distortion of ultrasonic waves may be micro-strain that evolves from the second phase and dislocations. In a solid that contains dislocations that are capable of glide displacements via small shear stresses, these dislocation displacements also effect the nonlinear stress-strain relation. Hikata et al. have completed a numerical as well as experimental investigation of the dislocation contribution to the generation of second harmonic ultrasonic waves in pure aluminum single crystal (Hikata et al., 1965). They also showed the model that the amplitude of the second harmonic wave is dependent on the dislocation displacement as follows:

$$A_2 \propto \frac{12}{5}\frac{\Omega \Lambda E^2 L^4 R^3}{\mu^3 b^2}\sigma \qquad (6)$$

where Ω is the conversion factor from shear strain to longitudinal strain, Λ is dislocation density, E is the second order elastic modulus, L is the dislocation loop length, R is the resolving shear factor, μ is the shear stress and σ is the static stress.

This indicates that the second harmonic wave is influenced on the dislocation displacement related to the dislocation density and loop length as well. In this study, the dominant microstructural development during fatigue of copper is the dislocation evolution such as dislocation density, loop length and substructures. In general, loop length of each dislocation may be changed depending on the state of dislocation pinning that is sometimes it may be increased or decreased. However, dislocation density of common metallic materials may be increased during the plastic deformation with about three orders or five orders of magnitude higher than the annealing state. Therefore, the dislocation density might be a dominant factor influencing the ultrasonic nonlinearity of copper during the fatigue.

5. Conclusion

The ultrasonic nonlinearity monotonically increased due to the fatigue driven plastic deformation. We have clearly seen that the dislocation density increased during fatigue of copper using the electron microscope and electron backscattered diffraction technique. In this work, we found that the change in dislocation density was enough to cause noticeable changes of the ultrasonic nonlinearity. Consequently, ultrasonic nonlinearity has been found to be sensitive to the dislocation behavior of copper during fatigue.

Acknowledgments

This work was supported by the Korea Science and Engineering Foundation (KOSEF) grant funded by the Korea government (MOST) (No.2007-00467)

References

Breazeale, M. A. and Thompson, D. O. (1963) Finite-Amplitude Ultrasonic Wave in Aluminum, Applied Physics Letters, Vol. 3, No. 5, pp. 77-78

Breazeale, M. A. and Ford, J. (1965) Ultrasonic Studies of the Nonlinear Behavior of Solids, Journal of Applied Physics, Vol.36, No. 11, pp. 3486-3490

Cantrell, J. H. (1994) Crystalline Structure and Symmetry Dependence of Acoustic Nonlinearity Parameter, Journal of Applied Physics, Vol. 76, No. 6, pp. 3372-3380

Cantrell, J. H. and Yost, W. T. (2000) Determination of Precipitate Nucleation and

Growth Rates from Ultrasonic Harmonic Generation, Applied Physics Letters, Vol. 77, No. 13, pp. 1952-1954

Cantrell, J. H. (2006) Dependence of Microelastic-Plastic Nonlinearity of Martensitic Stainless Steel on Fatigue Damage Accumulation, Journal of Applied Physics, Vol. 100, No. 6, pp. 0635081-0635087

Feltner, C. E. and Laird, C. (1967) Cyclic Stress-Strain Response of F.C.C. Metals and Alloys—II Dislocation Structures and Mechanisms, Acta Metallurgica, Vol. 15, pp. 1633-1653

Hertzber, R. W. (1976) *Deformation and Fracture Mechanics of Engineering Materials*, John Wiley & Son. Inc., NJ, pp. 415-464

Hikata, A., Chick, B. B. and Elbaum, C. (1963) Effect of Dislocations on Finite Amplitude Ultrasonic Waves in Aluminum, Applied Physics Letters, Vol. 3, No. 11, pp. 195-197

Hikata, A., Chick, B. B. and Elbaum, C. (1965) Dislocation Contribution to the Second Harmonic Generation of Ultrasonic Wave, Journal of Applied Physics, Vol. 36, No. 1, pp. 229-236

Hurley, D. C., Balzar, D., Purtscher, P. T. and Hollman, K. W. (1998) Nonlinear Ultrasonic Parameter in Quenched Martensite Steels, Journal of Applied Physics, Vol. 83, No. 9, pp. 4584-4588

Kim, C. S., Kim, Y. H. and Kim I. H. (2005) Ultrasonic Linear and Nonlinear Parameters in Cyclically Deformed Cu and Cu-35Zn Alloy, Key Engineering Materials, Vol. 297-300, No. 3, pp. 2134-2139

Kim, C. S. (2007) *Nondestructive Assessment of Microstructural Change by Fatigue and Creep*, Ph. D. Thesis, Korea University, Korea, pp. 46-77

Kim, C. S. and Park, I. K. (2008) Microstructural Degradation Assessment in Pressure Vessel Steel by Harmonistic Generation Technique, Journal of Nuclear Science and Technology, Vol. 45, No. 10, (in press)

Madhoun, Y., Mohamed, A. and Bassim, M. N. (2003) Cyclic Stress–Strain Response and Dislocation Structures in Polycrystalline Aluminum, Materials Science & Engineering, Vol. A359, pp. 220-227

McEvily. (1979) *Fatigue and Microstructure*, American Society for Metals, Metal Park, Ohio, pp. 149-203

Melngailis, J., Maradudin, A. A. and Seeger, A. (1963) Diffraction of Light by Ultrasound in Anharmonic Crystals, Physical Review, Vol. 131, No. 5, pp. 1972-1975

Meyendorf, N. G., Nagy, P. B. and Rockhlin, S. I. (2004) *Nondestructive Materials Characterization*, Springer Press, New York, pp. 206-232

Trivedi, P., Field, D. P. and Weiland, H. (2004) Alloying Effects on Dislocation Substructure Evolution of Aluminum Alloys, International Journal of Plasticity, Vol. 20, No. 3, pp. 459-476

Wallace, D. C. (1967) Thermoelasticity of Stressed Materials and Comparison of Various Elastic Constants, Physical Review, Vol. 162, No. 3, pp. 776-789