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Long-Term Aging Diagnosis of Rotor Steel Using Acoustic Nonlinearity

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Abstract The long-term aging of ferritic 2.25CrMo steel was characterized using the acoustic nonlinear effect in order to apply to diagnose the degradation behavior of structural materials. We measured the acoustic nonlinearity parameter for each thermally aged specimen by the higher harmonic-generation technique. The acoustic nonlinearity parameter increased with aging time due to equilibrium M_6C carbide precipitation, and has a favorable linear relation with Rockwell hardness. This study suggests that acoustic nonlinearity testing may be applicable to diagnostics on strength degradation in rotor steels.

Keywords: Acoustic Nonlinearity, Long-Term Aging, Harmonic Generation, Precipitate

1. Introduction

Material degradation shortens the lifespan of plant facility components under high temperature, and can lead to large-scale disasters due to sudden failures. The damages to the structures at nuclear power plants could possibly be the cause of a radiation leak. Therefore, safety evaluation or life assessment of such structures is considerably important and necessary. The in-service inspections are required to periodically evaluate safety and prepare for the following contingencies: Keeping recent facilities in operation?, Changing recent facilities?, If usi ng existing facilities, when/where do they need to be repaired?, If the change is required when would this be preferable?. Especially, the most common forms of damage caused to structural materials on prolonged exposure to high temperature and pressure are known to be fatigue, creep and long-term aging. Thus, it can be said that the assessment of material degradation are of utmost importance when checking the structural safety of a construction.

Evaluating the extent of structural damage, nondestructive evaluation(NDE) techniques have great potential to characterize material degradation over traditional techniques such as mechanical tests and microscopic observation. However, existing NDE techniques[1-4] have limitations in their depth of penetration, area of measurement and ability to detect microstructural damage. For these reasons, acoustic nonlinearity based technique[5-9] have been found to be very attractive to evaluation of bulk materials microstructure, possibly allowing evaluation of critical components during use and without removal from service.

The higher harmonic waves were typically generated by the distortion of the ultrasonic wave during propagation in the nonlinear solid, a phenomenon that is related to anharmonicity

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and strain of crystal lattices. Recently, Cantrell and Yost[10] presented a model in which acoustic harmonic-generation in aluminum depended on lattice coherency strains at the interface between the matrix and the dispersed phase. Hurley et al.[11] reported that the acoustic nonlinearity linearly increases with inhomogeneous strain due to precipitate generation in low alloy steel. Despite the recognized potential of acoustic nonlinearity, there are few examples of its use to characterize material damages of structural materials. In particular, there have been only a few reports of using acoustic nonlinearity to characterize long-term aging of high-temperature nuclear materials.

In the present study we assessed the thermal degradation(long-term aging) of typical rotor steel using acoustic nonlinearity, Rockwell hardness, and transmission electron microscopy (TEM) as part of a structural health monitoring (SHM) program.

2. Nonlinear Ultrasonic

Higher harmonic generation is phenomenologically related to nonlinearity in the elastic behavior of materials. The simple one-dimensional nonlinear stress-strain (σ-ε) relationship is expressed as $\sigma = E\varepsilon - (1/2)\beta\varepsilon^2$ using a second-order elastic constant (E, Young's modulus for uni-axial tension) and a second-order nonlinear elastic coefficient (β). If we consider a single frequency ultrasonic wave incident upon one end of a thin circular rod detected on the other end, then the differential equation representing motion for a longitudinal wave in thin circular rods can be obtained and a nonlinear wave equation can be expressed up to a second-order term as follows[12]:

$$\frac{\partial^2 u}{\partial t^2} = c_0^2 (1 - \beta \frac{\partial u}{\partial x}) \frac{\partial^2 u}{\partial x^2} \tag{1}$$

where $c_0 = \sqrt{(E/\rho)}$ is the longitudinal wave velocity, E is Young's modulus and β is nonlinearity parameter, u is the displacement in the x-direction, t is time and ρ is mass density of the solid.

An approximate solution of eqn. (1) is $u = u_0 + u_1$ where u_0 is a solution with $\beta = 0$, then the solution of eqn. (1) can be obtained by an iterative process [13]:

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

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

3. Experimental Details

A ferritic 2.25CrMo steel plate with the chemical composition shown in Table 1 was normalized at 900°C for 1 hour, then tempered at 720°C for 1 hour. Accelerated thermal degradation was performed at 630°C and interrupted at several predetermined times to observe different levels of damage. It is very difficult to know the actual in-service time to obtain the microstructure similar to that in-service. Researchers have used the self-diffusion equation of iron to obtain the equivalent aging time in ferritic steels. This technique is very reasonable and verified by

Table 1 Chemical composition of ferritic 2.25CrMo rotor steel (wt.%)

C	Si	Mn	Ni	Cr	Mo	P	Al	Fe
0.138	0.142	0.46	0.17	2.27	0.97	0.014	0.007	Bal.

some researcher though the equation is based on the self diffusion of iron[14]. The maximum aging time of 4800 hours at 630℃ was calculated to be the simulated service time of 260,000 hours at 538℃.

TEM specimens were prepared by electro-polishing using 10% perchloric acid and 90% glacial acetic acid. The energy dispersive spectroscopy analysis was used to identify the chemical composition of carbides. At the various degradation time points, Rockwell hardness (HRB) test was performed to measure strength degeneration due to thermal degradation.

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[8,9]. 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 nonlinearity parameter is determined from the eqn. (2) as follows:

$$\beta = \frac{8A_2}{A_1^2 k^2 x} \tag{3}$$

where x is the propagation distance, A_1 is the particle displacement amplitude of the fundamental wave and A_2 is the particle displacement amplitude of the secondary harmonic wave.

However, in this study we measured the relative nonlinearity parameter β’ that is proportional to the absolute nonlinearity parameter β. Since the relative value of β’ is enough to evaluate the relative changes of mechanical strength(hardness) during thermal aging. Most of all, β’ is easy to measure with a simple experimental set-up. The waveform was digitally processed using a power spectrum fast Fourier transformation in order to obtain the amplitudes, A_1 at the fundamental frequency and A_2 at the second harmonic frequency. The ultrasonic nonlinearity parameter β can usually be expressed in terms of the measured amplitude of the fundamental (A_1) and the second harmonic waves (A_2). In this study, we define and use a relative ultrasonic nonlinearity parameter $\beta' = A_2/A_1^2$ which is proportional to the absolute nonlinearity. Finally, we measured the normalized acoustic nonlinearity parameter (β'/β_0') where β’ is the nonlinearity parameter of a degraded specimen and β_0' is the nonlinearity parameter of an as-tempered specimen. As for the ultrasonic nonlinearity test specimen, the test materials were machined from a rectangular shape measuring 90 mm long, 20 mm wide and 20 mm thick.

4. Results and Discussion

Typical TEM micrographs of an as-tempered and 3700 h degraded specimen are shown in Fig. 1. Fig. 1(a) reveals numerous acicular carbides each 0.3 μm long. These were identified as M_2C carbides (Mo_2C) using energy dispersive spectroscopy analysis. Grain boundary (G.B.) precipitates were identified as rectangular parallelepiped $M_{23}C_6$ carbides ($Cr_{23}C_6$). Fig. 1(b) is a bright field image of a grain interior revealing only globular M_6C carbide (Mo_6C) precipitates without any fine acicular M_2C carbides. This M_2C carbide can not be observed after 1000 h of aging time. Fine acicular carbide particles might be dissolved in the matrix, and

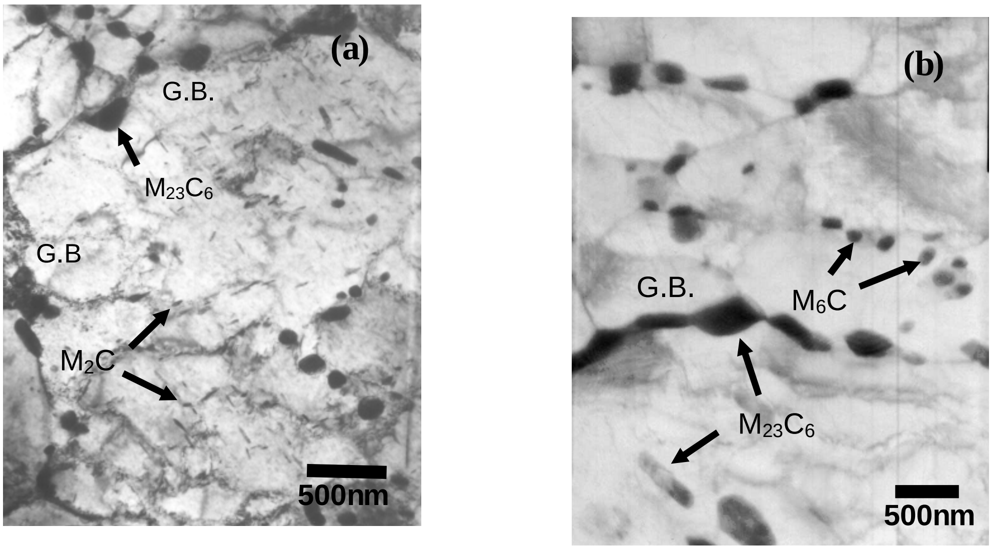

Fig. 1 TEM images showing carbide morphologies in ferritic 2.25CrMo steel: (a) as-tempered and (b) after 3700 h aging

could have transformed into the spherical equilibrium M_6C carbide phase during thermal degradation. Comparing several carbide morphologies, carbides have distinct morphologies. For instance, the M_2C phase is fine acicular carbide, $M_{23}C_6$ is a rectangular parallelepiped, and M_6C is globular[15-17]. The average sizes of M_6C, $M_{23}C_6$, and M_2C carbides are 0.11, 0.13 and 0.03 μm, respectively. All three carbides coarsen with increasing degradation time.

Fig. 2 shows the variation in HRB hardness and acoustic nonlinearity as a function of degradation time. The initial hardness after normalizing and tempering is depended on the M_2C carbide and dislocation density. The dislocation is one of the dominant contributions to the mechanical strength of ferritic steel. In addition, during the tempering and aging the grains are recovered resulting in decrease in dislocation density.

It can be observed from Fig. 2 that the hardness drops rapidly after short-time aging, which is attributed to the reduction in dislocation density due to dislocation annihilation[18,19] and dissolution of coherency M_2C carbide[15,20]. They have reported that the dislocation density decreased rapidly in early

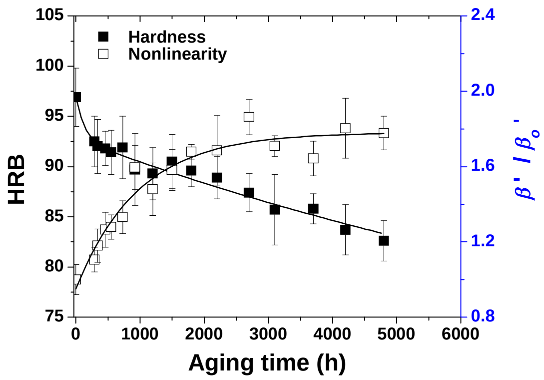

Fig. 2 Variation in hardness and acoustic nonlinearity as a function of aging time

time and changed little thereafter. For the early stage of aging of ferritic steel, the decrease in dislocation density is one of the reasons of softening of mechanical strength. However, the softening of mechanical strength of this kind of ferritic steels has been well known phenomenon of precipitate transformation[21-24] during total creep and thermal aging time. M_2C is believed to be completely coherent when first formed which is corresponding to the maximum hardness. However, the M_2C grows into well defined flattened needles, it is believed to have lost some of its coherency with the matrix. The mechanical strength of this kind of ferritic steel is well known to the prepitate (carbide) hardening and solid solution hardening mechanism [25]. For further aging time, the hardness monotonically decreased due to carbide coarsening. Since the carbide size is related to interparticle spacing, and hence to the resistance to deformation, the carbide has an major influence on hardness during thermal aging.

The normalized acoustic nonlinearity parameter (β'/β_0') increased during initial thermal degradation. Microstructural degradation affects the amplitude of second harmonic wave increase, and therefore, ultrasonic nonlinearity has been found to depend on microstructural degradation.

It has been assumed that the nonlinear stress-strain relation for the pure longitudinal wave can be represented by the power series [13]. The higher order terms of the equation are the cause of the generation of higher harmonics of a pure longitudinal wave as it propagates through the materials. From this nonlinear relation, the generalized Hook's law can be written as:

$$\sigma = E\varepsilon - \frac{1}{2}\beta\varepsilon^2 \tag{4}$$

where σ is the stress, $\varepsilon = \partial u / \partial x$ is the displacement gradient. In physical point of view, the nonlinearity parameter β is the amount of wave distortion passing through a solid with respect to the nonlinearity of the solid. Therefore, the nonlinearity parameter can be expressed using the second-order elastic constant (C_{11}) and the third-order elastic constants (C_{111}) as $\beta = -(3 + C_{111}/C_{11})$.

Hikata et al. [26] have been reported a model that the fraction change in the acoustic nonlinearity parameter ($\Delta\beta/\beta_0$) is given by

$$\Delta\beta / \beta_0 = \frac{24}{5}\frac{\Omega\Lambda L^4 R^3 {C_{11}}^2}{\beta_0 G^3 b^2}|\bar{\sigma}| \tag{5}$$

where $\Delta\beta = \beta - \beta_0$ is the change in the nonlinearity parameter, β_0 is the initial value, Ω is the conversion factor from shear stain to longitudinal strain, Λ is the dislocation density, L is a half of the average distance between the pinning points of the bowed dislocation segments, G is the shear modulus, b is Burgers vector, and $\bar{\sigma}$ is the local precipitate-matrix coherency stresses as $\bar{\sigma} = -32L^{-3}G\delta\bar{r}^3$ where δ is the precipitate-matrix lattice misfit parameter and $\bar{r}$ is the radius of precipitate.

The dominant microstructural change in this study is the generation and coarsening of precipitates. Precipitates have different crystalline lattice spacing from the matrix, which leads to a lattice mismatch measured by a misfit parameter as follows;

$$\delta = \frac{2(a_p - a_m)}{(a_p + a_m)} \tag{6}$$

where a_p and a_m are the lattice parameter for the precipitate and the matrix, respectively. The lattice parameters of M_6C and M_2C are 11.5 Å and 2.88 Å, respectively [27].

As already mentioned, M_6C carbide formation and growth are the dominant microstructural evolution events during thermal degradation. Fine M_6C carbide particles may produce a misfit strain between the precipitates and the matrix during thermal degradation. As more M_6C carbide particles are generated during thermal degradation, the net strain increases and distorts ultrasonic wave propagation through the solid.

Fig. 3 demonstrates the empirical correlation between hardness and acoustic nonlinearity during thermal degradation. Using statistical regression, a linear relationship (r^2=0.89, sd=1.8) was found between HRB hardness and the acoustic nonlinearity parameter (β'/β_0'). This means that acoustic nonlinearity may provide a reliable preliminary estimation of strength changes during long-term aging. This linear

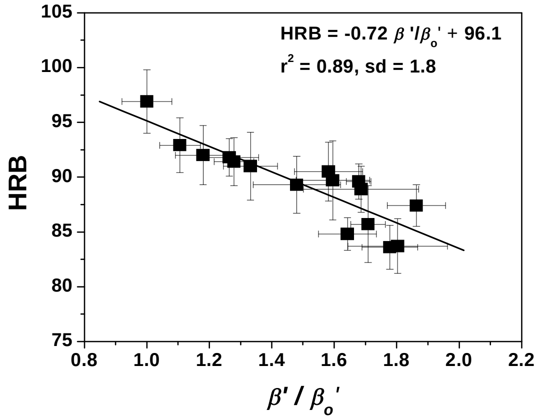

Fig. 3 Correlation between hardness and acoustic nonlinearity: The solid line represents linear least-square fit to the data. The correlation coefficient is 0.89 and stand deviation is 1.8

relation also suggests that the acoustic nonlinearity may be applied to degradation diagnostics on ferritic 2.25CrMo steels.

5. Conclusion

We have studied the application of acoustic nonlinearity to diagnose material degradation in a rotor steel. The hardness of ferritic 2.25CrMo steel decreased with the dissolution of M_2C carbide particles. These particles transformed to the equilibrium M_6C carbide phase, assumed a more spherical shape, and then became coarser during thermal degradation. The formation and growth of M_6C carbide particles produce a misfit strain between the precipitates and matrix, increasing the second harmonic component amplitude. A linear relationship between acoustic nonlinearity and hardness suggests that acoustic nonlinearity may be applied to diagnose long-term aging in rotor steels.

Acknowledgments

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