Roughness Estimation of Polycrystalline Iron Surface under High Temperature by Small Glancing Angle X-ray Scattering

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Characterization of polycrystalline iron surfaces before and after baking at 500°C for 20 h in a vacuum of \(10^{-6}\) Pa is performed by small glancing angle X-ray scattering with use of a compact ultrahigh vacuum (UHV) X-ray diffractometer. A broadening of the scattered X-ray intensity profiles at a small glancing angle incidence of the X-ray appears on the baked specimen. The experimental results are compared with calculated simulation with some surface structure models, and show that the roughness of the surface after the baking is estimated to be about ten times of that before baking. This result is consistent with the result of AFM observation, and shows that glancing angle X-ray scattering with the use of a compact UHV X-ray diffractometer is a useful technique for non-destructive inspection of industrial material surfaces.

KEY WORDS: surface roughness; polycrystalline iron; X-ray scattering.

1. Introduction

In steel, the need to have knowledge of surface and interfacial phenomena is almost axiomatic. For situations with not only degradation process such as corrosion, oxidation, wear, etc. but also surface treatment process such as descaling, conversion coating, electroplating, and so on, the interaction at the surface and interface are of prime importance. Therefore, numerous characterization techniques of surface and interface of steel have been proposed and used.

Glancing angle X-ray scattering is a useful technique for the study of surfaces and interfaces. With the development of synchrotron radiation (SR) facilities, many studies of surfaces and interfaces have been made using intense SR X-ray beams. This technique will also be applicable to surface characterization of industrial materials such as steel. For the surface X-ray scattering experiments to be performed in a small laboratory room, the present authors constructed a compact ultrahigh vacuum (UHV) X-ray diffractometer, in which an 18 kW rotating-anode X-ray source, an UHV specimen chamber and a two-circle diffractometer, were all arranged on a single optical table of 70 cm\(\times\)90 cm. The apparatus makes use of total reflection of X-rays and is designed in such a way that the atoms only at the surface, ca. 10 atomic layers deep, contributes to the scattering. With this small apparatus we performed characterization of polycrystalline iron surfaces before and after baking at 500°C for 20 h in a vacuum of \(10^{-6}\) Pa. Angular distributions of the scattered X-ray intensity from the surfaces were measured at several glancing angle incidences of the X-ray. The experimental results were compared with calculated simulation with some surface structure models, and the surface roughness was estimated. Based on these results, we proposed a method of surface characterization of steel as industrial materials.

2. Experimental

Details of the above compact UHV diffractometer are described elsewhere, and only a brief description of the present experimental arrangement is given here. Figure 1 shows a schematic top view of the apparatus. A Cu-K\(\alpha_1\) X-ray beam (wave length; 0.154 nm, energy; 8 keV) from an 18 kW rotating-anode source was well monochromatized and collimated through a germanium crystal monochromator as are shown at the left of Fig. 1. The cross section of the X-ray beam was 0.05 mm wide and 0.1 mm high. The X-ray beam was then introduced into the UHV chamber through a Be window, 0.2 mm thick and 20 mm in diameter. The UHV chamber and a scintillation counter were mounted on a two-circle \(\theta_i-\theta_s\) diffractometer whose rotating axes were both normal to the plane of the figure. A surface specimen in the UHV chamber was placed with its surface standing normal to the figure plane. The specimen holder had a heater to anneal the specimen in the UHV, which could heat the specimen up to 500°C. The X-ray beam was then incident upon the specimen surface at a glancing angle \(\theta_i\), as is shown in Fig. 2. Divergence of the incident beam was about 0.3 mrad in full width at its half maximum, and its maximum intensity was 17 000 counts per second when the power of the Cu rotating-anode source was 50 kV–300 mA. Scattered X-ray beams from the speci-
men surface in the UHV chamber were taken out through another Be window, limited by the other slits, and then received by a scintillation counter as shown at the right of Fig. 2. The acceptance angle of the scattered X-ray beams limited by the slits was 0.3 mrad in the angular direction $q_s$.

Measurements of scattered X-ray intensities were performed on mechanically polished pure iron polycrystalline surfaces (ferrite, 3 nines purity) of $10^{10}/H1100310$ mm$^2$. The surfaces of the iron samples were mechanically polished by diamond powder of 0.5 micrometer diameter at Materials Research Laboratory of Kobe Steel Ltd. One of the surface X-ray scattering experiments was performed on the iron surface at room temperature, at about 20°C, in a vacuum of $10^{-6}$ Pa. Other was performed after baking at 500°C for 20 h in a vacuum of $10^{-6}$ Pa.

3. Results and Discussion

The results of the scattering experiments on the iron surface at 20°C are shown in Fig. 2(a). Each of the $\theta_s$ distributions of the scattered X-ray intensity has a sharp peak at the respective specular reflection angle of $\theta_s=2\theta_i$. The width of the specular peak becomes slightly broader with increase...
of the glancing angle. These angular distributions include information about the surface roughness. If the surface is assumed to be flat and infinite in extent, the specular peak is $\delta$ function. When the flat surface has finite-size, the peak is broadened by an amount inversely related to the size of the flat surface.

Figure 2(b) shows the angular distributions of the scattered X-ray on the iron surface at which the temperature was maintained at 500°C during measurement after baking at 500°C for 20 h in a vacuum of $10^{-6}$ Pa. Although each of these angular distributions also has a peak at the respective specular reflection angle, these peaks are broad and have small intensity maximum when compared with those of Fig. 2(a). The results of Figs. 2(a) and 2(b) can provide some possibilities to estimate the roughness of the polycrystalline iron surfaces. When the surface roughness is large, the specular peak will become low and broad. Then the results of Fig. 2(b) show that the iron surface roughness became large after baking at 500°C. In the measurement on the iron surface at room temperature after one day of the baking at 500°C also, the broadness of the angular distributions did not change from those on the surface at 500°C. This shows that Debye–Waller factor does not affect this broadness of the angular distribution.

We also observed the same surface before and after baking by atomic force microscopy (AFM). Figure 3(a) shows the AFM image and the roughness profiles of polished polycrystalline iron surface before the baking at room temperature, and Fig. 3(b) shows those of the iron surface at room temperature after a few days of baking at 500°C. Enhancement of the surface roughness was clearly observed on the baked iron surface. The r.m.s. roughness of the polished iron surface in Fig. 3(a) was 2.6 nm, and that of the baked iron surface in Fig. 3(b) was 22.7 nm. AFM observation shows that the surface roughness was enhanced about ten times by the baking.

We have estimated the roughness by analyzing the angular distributions of scattered X-ray intensities, together with the observations by AFM. Field amplitude $E_{sc}$ of X-ray scattered on a surface can be calculated using the Distorted-wave Born Approximation (DWBA) as the following Eq. (1),

\[ E_{sc} = \frac{2i}{\lambda} \int \frac{d^2 R}{4\pi} \left( \frac{\sin k_{sc} R}{k_{sc} R} \right) g(R) e^{-i k_{sc} R} \]

This equation represents the amplitude of the scattered wave, where $k_{sc}$ is the scattering wavevector, $\lambda$ is the wavelength of the X-ray, and $g(R)$ is the electron density at a distance $R$ from the surface.
where

\[
E_{sc} = \int_{0,1}^{0,1} \exp \left[ i (k - k_0) \cdot r \right] dr \tag{1}
\]

Here \(k_0\) and \(k\) are the wave vectors of the incident and scattered X-rays. Integral calculus is performed over the surface plane. As a simulation model of rough surface we supposed that it is made up of the small tilted planes as is shown in Fig. 4. First, integral calculus is performed over each small plane, and the field amplitude \(E_{sc}\) of X-ray scattered on the small flat plane is derived respectively. Next, those results are added about the small planes with finite length and tilt distribution as is shown in Fig. 5, where length \(l\) of a plane with tilt angle \(\alpha\) is

\[
l = L \cdot \exp \left( - \frac{\alpha^2}{2\phi^2} \right) \tag{2}
\]

The r.m.s. roughness of this surface model is about \(0.58L\phi\), where \(L\) and \(\phi\) are defined in Eq. (2) and in Fig. 5. With this surface model we calculated the angular distribution of the scattered X-ray intensity on a rough surface. Figure 6(a) shows calculated distributions of the scattered X-ray intensity on the surface made up of small tilted planes with length \(l=32 \mu m\) and \(\phi=0.01^\circ\) in Eq. (2). These values of \(l\) and \(\phi\) are derived so that the full width at half maximum (FWHM) of the peak at specular angle in the calculated angular distribution might correspond to the experimental result of the angular distribution of the scattered X-ray intensity on the polished iron surface. Figure 6(b) shows calculated distributions on the surface made up of small tilted planes with length \(l=25 \mu m\) and \(\phi=0.1^\circ\). These values of \(l\) and \(\phi\) are derived so that FWHM of the specular peak in the calculated angular distribution might correspond to the experimental result on the baked iron surface.

Then it is shown that the glancing angle X-ray scattering can lead the angular distribution of the scattered X-ray intensity to estimate surface roughness. The roughness of the model surface with tilt distribution \(\phi=0.01^\circ\) is 3.2 nm in Fig. 6(a) and the model surface with tilt distribution \(\phi=0.1^\circ\) is 25.3 nm in Fig. 6(b). These results are shown in Table 1.

These results of surface roughness 3.2 nm and 25.3 nm estimated from the X-ray scattering correspond to the roughness 2.6 nm of polished surface and the roughness...
22.7 nm of baked surface by AFM observation. The roughness of two surfaces derived by the X-ray scattering was estimated to have about ten times the difference, which is consistent with the results of AFM observations.

The structures in the direction parallel to the surface plane estimated by X-ray scattering were different with AFM observations. The length of small planes estimated by X-ray scattering was order of 10 μm, while the structural dimension in the surface by AFM observation was order of 1 μm. Thus the structure in the direction parallel to the surface plane was not estimated precisely in the present X-ray scattering experiment.

4. Summary

Small glancing angle X-ray scatterings on polycrystalline iron surfaces before and after baking at 500°C were performed with use of a compact UHV X-ray diffractometer. A broadening of the scattered X-ray intensity profiles appeared on the baked specimen. Analyzing with calculated simulation it was found that the broadening is due to the enhancement of surface roughness, and the order degree magnitude of surface roughness was estimated. This result was consistent with the result of AFM observation, and shows that glancing angle X-ray scattering with the use of a compact UHV X-ray diffractometer is a useful technique for non-destructive estimation of industrial material surfaces.

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