Thermal Radiation Spectroscopy Diagnosis for Temperature and Microstructure of Surfaces*

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Surface microstates of real surfaces of solid materials change in industrial environments, or they are changed in industrial surface processes positively. A real time diagnosis technique for the surface microstates should be developed for the process control. We propose a diagnosis technique for temperature and microstructure of real surfaces on a basis of the hardware performance of our wide-spectral-range high-speed spectrophotometer system. With respect to the temperature, an active spectral pyrometer technique is adopted, in which the reflection of the surface is measured as well as the self-emission. With respect to the microstructure, an attention is paid to the interference and diffraction of radiation in a new real surface model. An experiment is made on a metal surface in a high-temperature air-oxidation process to verify the performance of this technique. Spectra of emission and reflection energy of radiation in a near-infrared through infrared region are measured at every 2 s, and analyzed. Time transition of the surface temperature of an order of 1 100 K is estimated within an inaccuracy of 10 K. The average thickness d and rms roughness σ_1 of the surface film are estimated to be in regions of $d=0.08\sim 3.8\ \mu\text{m}$ and $\sigma_1=0.01\sim 0.71\ \mu\text{m}$, respectively.

Key Words: Thermal Radiation, Spectroscopic Measurement, Radiation Pyrometry, Surface Diagnosis, Real Surface, Surface Film, Surface Roughness

1. Introduction

Real surfaces of solid materials are rough and covered by any surface films. The microstructure can change drastically in crude industrial environments. The surface microstructure can be processed positively to realize various functions on the surface. Such change in the surface microstructure affects the spectral characteristics of thermal radiation of the surfaces sensitively and extensively.

Authors have developed a wide-spectral-range high-speed spectrophotometer system⁽¹⁾ for a study on thermal radiation phenomena⁽²⁾. The system measures the transition of spectra of reflection and emission of radiation in a visible through infrared region of wavelength $0.30\sim 11\ \mu\text{m}$ with a cycle time of 2 s. By using this system we have investigated the dynamics

of spectrum transition on metallic materials in processes of physical/chemical reactions. It was found that measured spectral phenomena reflect the change in surface microstructure sensitively. Considering the sensitive dependence of radiation characteristics on the surface microstructure, a new technique is expected to be developed for the remote and real time diagnosis of microstructure of real surfaces. On the other hand, development of an accurate radiation pyrometry for real surfaces is another important target of thermal engineering. The technique is highly precise but not accurate in the most cases when the radiation characteristic value like the emittance of the surface is unknown or changing as in the above case. Thermal radiation pyrometry which measures the change in radiation characteristics and temperature simultaneously should be established. Spectral pyrometry technique⁽³⁾⁻⁽⁵⁾ is a promising one for this purpose. In 1999, we presented the first trial of a spectral diagnosis technique which combines the temperature measurement and the surface microstructure measurement⁽⁶⁾.

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In the present work, on a basis of the hardware of our improved spectrophotometer system⁽¹⁾, we propose an improved version of algorithm for the spectral surface diagnosis of temperature and microstructure of real surfaces. The algorithm is backed up by ideas of heat transfer science and film optics. In order to demonstrate the performance of the algorithm, we make an experimental examination to measure the spectra of emission and reflection of radiation on a metal surface in a high-temperature air-oxidation process. The results are analyzed to estimate the transition of surface temperature T^{opt} , spectra of emittance ϵ_N^{opt} and reflectance R_{NN} of the surface, reflectance ratio β of the surface, average thickness d , and roughness σ_i of the surface film. The proposed technique is promising for the in-process diagnosis for the quality and safety control in materials and/or energy production processes.

Nomenclature

A	: area of measured surface, m^2
d	: film thickness, m
f	: probability density distribution functions of the normal distribution, m^{-1}
I	: intensity of radiation, $\text{W}\cdot\text{m}^{-2}\cdot\text{sr}^{-1}$ or $\text{W}\cdot\text{m}^{-2}\cdot\text{sr}^{-1}$
i	: imaginary unit
k	: wave number of radiation in vacuum, $=2\pi/\lambda$, m^{-1}
	imaginary part of optical constant $\hat{n}$
n	: index of refraction
$\hat{n}$	: optical constant, $=n+ik$
q	: radiation flux, $\text{W}\cdot\text{m}^{-2}$
R	: (energy) reflectance
$\hat{r}$	: Fresnel's complex coefficients of reflection
T	: temperature, K
t	: time, s
$\hat{t}$	: Fresnel's complex coefficients of transmission
u	: measured emission energy, V
v	: measured reflection energy, V
z	: coordinate, m
β	: reflection ratio
ϵ	: (directional) reflection
θ	: zenith angle, rad
λ	: wavelength of radiation in vacuum, m
λ_i	: wavelength point (wavelength region) i , m
σ	: rms roughness of interface, m
Ω	: solid angle, sr
$\langle \rangle$	: expectation
$\hat{}$	: complex quantity
Subscripts and Superscripts	
B	: black body
H	: hemispherical

i	: wavelength point (wavelength region) i
i, j, k	: media i, j and k
N	: normal
opt	: optical diagnosis
R	: reflection
RF	: rough film model
s, p	: s-polarized and p-polarized radiation
SF	: specular film model
sys	: surface system, (substrate) + (film)
tc	: measured by thermocouple
0	: incident, air
1, 2	: film, substrate
I, II	: air-film interface, film-substrate interface

2. Algorithm for Real Surface Diagnosis

2.1 Ideas for diagnosis algorithm

We propose an algorithm for real surface diagnosis on a basis of the developed spectrophotometer system⁽¹⁾. In this system, radiation is dispersed as spectral radiation by diffraction gratings and detected by linear arrays of optical detector elements. In the present work, we measure the spectra of reflection and emission of radiation at a number of wavelength points in a near-infrared through infrared region simultaneously and repeatedly with a cycle time of 2 s.

Ideas for the algorithm of the surface diagnosis are as follows. With respect to the temperature, first, an idea of spectral pyrometer is adopted to deal with radiation information at a number of wavelengths. Second, an active technique is adopted, in which the target surface is irradiated and reflection is measured as well as the self-emission of the surface. Third, an energy ratio of the specular-hemispherical reflection at the rough real surface is dealt from a hemispherical point of view in heat transfer science. With respect to the microstructure, first, a simple and non-sophisticated optical/surface model is presented for the surface diagnosis. Second, a special attention is paid to the minima and maxima of radiation interference at the rough surface with a film to estimate the thickness of the surface film. Third, another attention is paid to the diffraction of radiation into a particular direction to estimate the roughness of the surface film.

2.2 Outline of surface diagnosis

The algorithm of real surface diagnosis proposed in this work is applicable typically to that for a surface whose surface state is changing, e.g. a metal surface in a high-temperature air-oxidation process⁽¹⁾. Such a surface consists of a substrate opaque for radiation and a single-layer film on the substrate. The temperature of such a surface system is assumed to be homogeneous over the surface system. The optical constant spectrum of the surface film is assumed to be constant in the depth direction and over the tempera-

ture range considered. The film thickness and roughness of the upper and lower interfaces of the film increase with time. At the initial stage, the surface can already be rough and covered by a film which is far thinner than the wavelength of radiation considered. Spectral radiation of wavelength point λ_i which is covered by each of the detector elements in the detector arrays is that of a narrow band wavelength region of $(\lambda_i - \Delta\lambda_i/2) \leq \lambda \leq (\lambda_i + \Delta\lambda_i/2)$.

Figure 1 explains the outline of the diagnosis. At each time t we measure spectra of emission energy $u_i(t)$ in the direction of a zenithal angle θ_0 and reflection energy $v_i(t)$ in the same direction for the incidence of a zenithal angle $(-\theta_0)$ over the wavelength points λ_i ($i=1, 2, \dots, n$). The angle θ_0 is set actually to be near-normal to the surface, and the emittance ϵ_N and reflectance R_{NN} or R_{NH} in the following description accompany a subscript N, which stands for 'normal'. The spectra of $u_i(t)$ and $v_i(t)$ are input quantities to the diagnosis system. These quantities are analyzed. The output quantities are the followings: first, on the real surface system, temperature $T(t)$, spectrum of emittance $\epsilon_{Ni}(t)$ in the direction of θ_0 , spectrum of reflectance $R_{NNi}(t)$ in the direction of θ_0 for the incidence of angle $(-\theta_0)$ and reflectance ratio $\beta(t)$ defined in Eq. (12); second, on the surface microstructure, average thickness $d(t)$ of the surface film and root-mean-squares (rms) roughness $\sigma_I(t)$ and $\sigma_{II}(t)$ of the upper and lower interfaces of the film. We call the above quantities "temperature and microstructure" of real surface, and estimate

Fig. 1 Outline of the diagnosis for temperature and microstructure of a real surface

them at each time t .

2.3 Measured emission and reflection energies

Figure 2 explains the angular configuration of the measurement of emission and reflection of radiation in the measuring system postulated⁽¹⁾. At each time t , measured emission energy $u_i(t)$ at each wavelength point λ_i , which has a unit of V, is described by,

$$u_i(t) = \epsilon_{Ni}(t) \cdot P_i\{T(t)\} \quad (1)$$

$$P_i(T) = \alpha \cdot I_{Bi}(T) \cdot \Delta A \cdot \cos \theta_0 \cdot \Delta \Omega \quad (2)$$

$$I_{Bi}(T) = \int_{\lambda_i - \Delta\lambda_i/2}^{\lambda_i + \Delta\lambda_i/2} I_{B\lambda}(T) d\lambda \quad (3)$$

where $I_{B\lambda}(T)$ [W/(m²·sr)] is spectral radiation intensity at wavelength λ of the blackbody at temperature $T(t)$, and $I_{Bi}(T)$ [W/(m²·sr)] is radiation intensity of wavelength point (narrow band wavelength region) λ_i of the blackbody at temperature $T(t)$. ΔA and $\Delta \Omega$ are area and solid angle of radiation of the measured surface, respectively. α is a conversion efficiency of the spectroscopic/electronic sub-systems of the measuring system which is assumed to be constant over the wavelength region of λ_i . The overall system constant $P_i(T)$ for the emission measurement is calibrated at each wavelength point λ_i as a function of temperature T , by an emission measurement on a blackbody setup of the area $\Delta A \cdot \cos \theta_0$ whose normal emittance is known.

In the reflection measurement at each wavelength point λ_i , radiation energy fluxes of incidence and reflection q_{oi} and $q_{ri}(t)$ are described, respectively, by,

$$q_{oi} = I_{oi} \cdot \cos(-\theta_0) \cdot \Delta \Omega_0 \quad (4)$$

$$q_{ri}(t) = I_{ri}(t) \cdot \cos \theta_0 \cdot \Delta \Omega \quad (5)$$

where I_{oi} and $I_{ri}(t)$ are intensity of radiation of incidence and reflection, respectively, and $\Delta \Omega_0$ is solid angle of the incidence. The solid angle of measured reflection is the same as the angle $\Delta \Omega$ for the measured emission. In our measurement system, the optical sub-systems for incidence and reflection are designed to be symmetric,

$$\cos(-\theta_0) \cdot \Delta \Omega_0 = \cos \theta_0 \cdot \Delta \Omega \quad (6)$$

Energy reflectance $R_{NNi}(t)$ to the solid angle $\Delta \Omega$ in the direction of θ_0 is equal to the intensity reflectance $I_{ri}(t)/I_{oi}$.

$$R_{NNi}(t) = q_{ri}(t)/q_{oi} = I_{ri}(t)/I_{oi} \quad (7)$$

At each time t , measured reflection energy $v_i(t)$,

Fig. 2 Angular configuration of measured emission and reflection of radiation

which has a unit of V, is described by,

$$v_i(t) = R_{NNi}(t) \cdot Q_i \cdot \cos \theta_0 \cdot \Delta\Omega \quad (8)$$

$$Q_i = \alpha_i \cdot I_{0i} \cdot \Delta A \quad (9)$$

The overall system constant Q_i for the reflection measurement is calibrated at each wavelength point λ_i , by a reflection measurement on an optically smooth surface whose optical constant is known.

Microstate of the real surface changes with time t , and emittance ε_{Ni} and reflectance R_{NNi} of the surface depend on λ_i and T . Temperature T changes with time t . Accordingly, u_i , v_i , ε_{Ni} and R_{NNi} are functions of time t , wavelength λ_i and temperature T . In the following description, however, notation (t) for time t and subscript i for wavelength "point (narrow band wavelength region)" λ_i are abbreviated for simplicity.

2.4 Diagnosis of temperature and emittance spectra of surface system

Radiation properties of the real surface system are assumed to be isotropic in the azimuthal direction. In this case, in Fig. 2, the emittance in the direction of $(-\theta_0)$ and the emittance in the direction of θ_0 are equal to each other. Among the emittances $\varepsilon_N(-\theta_0)$, $\varepsilon_N(\theta_0)$ and hemispherical reflectance R_{NH} for incidence in the direction of $(-\theta_0)$, the Kirchhoff's law⁽⁷⁾ is held rigorously.

$$\varepsilon_N(-\theta_0) + R_{NH} = \varepsilon_N(\theta_0) + R_{NH} = 1 \quad (10)$$

The hemispherical reflectance R_{NH} is described by,

$$R_{NH} = \int_{\text{hemisphere}} R \cdot \cos \theta \cdot d\Omega \quad (11)$$

The real surface reflects radiation imperfectly-diffusely and the optical detector element measures a part of this reflection. We pay an attention to the ratio β of the measured reflection energy to the hemispherical reflection energy,

$$\begin{aligned} \beta &= R_{NN} \cdot I_0 \cdot \Delta A \cdot \cos \theta_0 \cdot \Delta\Omega \\ &\quad / \int_{\text{hemisphere}} R_{NN} \cdot I_0 \cdot \Delta A \cdot \cos \theta \cdot d\Omega \\ &= R_{NN} \cdot \cos \theta_0 \cdot \Delta\Omega / R_{NH} \end{aligned} \quad (12)$$

and describe the ratio β by a polynomial expansion form of wavelength λ ,

$$1/\beta = \sum_{k=0}^m a_k \lambda^k \quad (13)$$

where β is a function of time t , temperature T and wavelength λ . Coefficients a_k are functions of t and T , and independent of λ . By summarizing the above, emission energy u and reflection energy v are correlated as follows:

$$u = \varepsilon_N \cdot P(T) \quad (14)$$

$$v = \beta \cdot (1 - \varepsilon_N) \cdot Q \quad (15)$$

At each time t , the total number of measured quantities u and v are $(2n)$ corresponding to the n wavelength points. On the other hand, the total number of unknown quantities to be analyzed in this diagnosis is $(n+m+2)$: temperature T , as many as n emittances

ε_N at n wavelength points, and as many as $(m+1)$ coefficients a_k for β spectrum. Accordingly, within an extent of $2n \geq n+m+2$, measured spectra of emission energy u and reflection energy v at each time t should be analyzed to determine temperature T and spectra of ε_N and β at each time t . The spectrum of R_{NN} is easily determined by Eq.(8).

2.5 Reflection model for a real surface

For the diagnosis of microstructure of real surfaces, a simple surface model is needed. Sophisticated models accompanying many model parameters⁽⁸⁾ do not fit this purpose of the diagnosis. We propose a model for describing radiation reflection at the rough real surface with a surface film for our diagnosis technique, in which we measure the reflection spectra in a specular direction of the target surface. Figure 3 explains the model. The surface system consists of an air-film interface I and a film-substrate interface II, which compose a single-layer film. Radiation wave is incident on the surface system. The wave is reflected multiply in the film. The superimposed and interfered wave of the elementary waves is observed by the detector sub-system. In fact, the interfaces I and II are not many finite size of elements as shown in Fig. 3, but they are flat interfaces parallel to each other and infinitely wide in the horizontal direction. The z -coordinates of the interfaces I and II are determined at every step of the multiple reflection by following probability rules. That is, the phases of the elementary waves of the multiple reflection are modified by weights of the probability distribution of the z -coordinates of the interfaces. In the measured radiation, which is the superimposition of the elementary waves, the sharpness of the interference decreases. Absorption of radiation in the film occurs in a similar manner as that in a simple parallel film.

We call the above surface model for a rough real surface with a film (Fig. 3) an RF model (rough film model). The surface model for a specular surface with a flat film, which is called an SF model (specular film model), is a special case of the RF model. The RF model does not consider the height distribution in the horizontal direction. Accordingly, it is natural that this model does not describe the overall view of directional characteristics of radiation diffraction or radiation scattering by the rough surface. But, the discussion up to Eq.(29) is exactly correct, so far as we consider the diffracted radiation in the specular direction. This fact can be understood if we learn a simple theory of diffraction by an optical grating, which fits the Kirchhoff's diffraction theory⁽⁹⁾. Reflected radiation wave observed in the specular direction is not affected by the height distribution in the horizontal direction, provided the inclination of

Fig. 3 Model of radiation reflection at a real surface with a surface film with rough interfaces

the rough surface elements is small so that the multiple diffraction or the shadow effect of radiation can be neglected, and provided the incidence and the corresponding specular reflection are not measured in a very oblique direction.

Consider the z -coordinates of interfaces I and II determined by a kind of probability density distribution functions $f[\text{m}^{-1}]$ of the normal distribution,

$$f_M(z) = 1/\{(2\pi)^{1/2}\sigma_M\} \exp\{-(z - z_{mM})^2/(2\sigma_M^2)\} \quad (16)$$

where the subscript M on f , z_m and σ stands for interfaces I and II. Coordinates z_{mI} and z_{mII} are the average positions of interfaces I and II, and the average thickness d of the film is,

$$d = z_{mII} - z_{mI} \quad (17)$$

Standard deviations σ_I and σ_{II} of the distributions of z_{mI} and z_{mII} correspond to the rms roughness of interfaces I and II, respectively. A plane wave whose wavelength and wave numbers in vacuum are λ and k ($=2\pi/\lambda$), respectively, is incident on this surface system, and multiple reflection occurs. The reflection phenomenon follows the Snell's formulae and the Fresnel's formulae.

In the case when the z -coordinates of interface I for the 0-th, 1st, 2nd, ... interface reflections and those of interface II for the 1st, 2nd, ... interface reflections are written by a particular set of $[z_{I0}, z_{I1}, z_{I2}, \dots, z_{I11}, z_{I12}, \dots]$, the complex amplitude reflectance $\hat{r}_{\text{sys}}$ of the surface system is described by,

$$\begin{aligned} \hat{r}_{\text{sys}} = & \hat{p}_0 \hat{r}_{01} \hat{p}_0 + \hat{p}_1 \hat{t}_{01} \hat{q}_{11} \hat{r}_{12} \hat{q}_{10} \hat{t}_{10} \hat{p}_0 + \\ & + \hat{p}_2 \hat{t}_{01} \hat{q}_{22} \hat{r}_{12} \hat{q}_{21} \hat{r}_{10} \hat{q}_{11} \hat{r}_{12} \hat{q}_{10} \hat{t}_{10} \hat{p}_0 + \\ & + \dots \end{aligned} \quad (18)$$

where $\hat{r}_{ij}$ and $\hat{t}_{ij}$ are, respectively, the Fresnel's complex coefficients of reflection and transmission in the case when radiation propagates from a medium i to another medium j . The Fresnel's coefficients $\hat{r}_{ij}$ and $\hat{t}_{ij}$ are functions of optical constants $\hat{n}_i$ and $\hat{n}_j$ of the media i and j , and $\hat{p}_i$ and $\hat{q}_{KL}$ are defined by,

$$\hat{p}_i = \exp(ikz_{iI} \cos \theta_0) \quad (19)$$

$$\hat{q}_{KL} = \exp\{ik(z_{iIK} - z_{iIL})\hat{n}_i \cos \hat{\theta}_i\} \quad (20)$$

where $\cos \hat{\theta}_i$ is calculated by the Snell's formulae. When distributions of z -coordinates $[z_{I0}, z_{I1}, z_{I2}, \dots, z_{I11}, z_{I12}, \dots]$ of interfaces I and II follow Eq. (16), then the expectation $\langle \hat{r}_{\text{sys}} \rangle$ of $\hat{r}_{\text{sys}}$ is written by,

$$\begin{aligned} \langle \hat{r}_{\text{sys}} \rangle = & \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \dots \int_{-\infty}^{+\infty} f_I(z_{I0}) f_I(z_{I1}) f_I(z_{I2}) \dots \\ & \cdot f_{II}(z_{II1}) f_{II}(z_{II2}) \dots \cdot \hat{r}_{\text{sys}} \cdot dz_{I0} dz_{I1} dz_{I2} \dots \\ & \cdot dz_{II1} dz_{II2} \dots \\ = & \hat{r}_{01} \hat{s}_0 + \hat{t}_{01} \hat{r}_{12} \hat{t}_{01} \hat{s}_1 \hat{s}_2 \hat{s}_1 + \\ & + \hat{t}_{01} \hat{r}_{12} \hat{r}_{10} \hat{r}_{12} \hat{t}_{10} \hat{s}_1 \hat{s}_2 \hat{s}_3 \hat{s}_1 + \dots \end{aligned} \quad (21)$$

where $\hat{s}_0$, $\hat{s}_1$, $\hat{s}_2$ and $\hat{s}_3$ are,

$$\hat{s}_0 = \int_{-\infty}^{+\infty} f_I(z) \exp(i2kz \cos \theta_0) dz \quad (22)$$

$$\hat{s}_1 = \int_{-\infty}^{+\infty} f_I(z) \exp\{ikz(\cos \theta_0 - \hat{n}_1 \cos \hat{\theta}_1)\} dz \quad (23)$$

$$\hat{s}_2 = \int_{-\infty}^{+\infty} f_{II}(z) \exp(i2kz \hat{n}_1 \cos \hat{\theta}_1) dz \quad (24)$$

$$\hat{s}_3 = \int_{-\infty}^{+\infty} f_I(z) \exp(-i2kz \hat{n}_1 \cos \hat{\theta}_1) dz \quad (25)$$

The values of the complex quantities can be calculated analytically by using an integration formula for the error function. The second and further terms in the right hand side of Eq. (21) mean the summation of an infinite geometrical series of the first term ($\hat{t}_{01} \hat{r}_{12} \hat{t}_{01} \hat{s}_1^2 \hat{s}_2$) and the common ratio ($\hat{r}_{12} \hat{r}_{10} \hat{s}_2 \hat{s}_3$). The Fresnel's complex coefficients in Eq. (21) follow the Stokes' relations,

$$\hat{r}_{ij} = -\hat{r}_{ji}, \quad \hat{t}_{ij} \hat{t}_{ji} = 1 - \hat{r}_{ij}^2 \quad (26)$$

Thus, the equation for $\langle \hat{r}_{\text{sys}} \rangle$ is summarized by,

$$\begin{aligned} \langle \hat{r}_{\text{sys}} \rangle = & \hat{r}_{01} \hat{s}_0 + \hat{t}_{01} \hat{r}_{12} \hat{t}_{01} \hat{s}_1^2 \hat{s}_2 / (1 - \hat{r}_{12} \hat{r}_{10} \hat{s}_2 \hat{s}_3) \\ = & \hat{r}_{01} \hat{s}_0 + (1 - \hat{r}_{01}) \hat{r}_{12} \hat{s}_1 \hat{s}_2 / (1 + \hat{r}_{01} \hat{r}_{12} \hat{s}_2 \hat{s}_3) \end{aligned} \quad (27)$$

The measured reflectance R_{NN} is the ratio of reflected energy to the energy of the wave of natural radiation incident on the surface system, and it is written as the average of values of $\langle \hat{r}_{\text{sys}} \rangle$ calculated for s- and p-polarized radiation.

$$R_{\text{NN}} = (|\langle \hat{r}_{\text{sys}} \rangle_s|^2 + |\langle \hat{r}_{\text{sys}} \rangle_p|^2) / 2 \quad (28)$$

Here, let the wavelength λ , and the angle ($-\theta_0$) of incident radiation, optical constants $\hat{n}_i$ and $\hat{n}_j$ of the film and the substrate are known, then the reflectance R_{NN} is a function of unknown variables d , σ_I and σ_{II} .

$$R_{\text{NN}} = R_{\text{NN}}(d, \sigma_I, \sigma_{II}) \quad (29)$$

The above discussion in Section 2.5 is for monochromatic radiation of wavelength λ . But, actually, measured radiation is of a wavelength point (a narrow band wavelength region) as mentioned in Section 2.2. In the present diagnosis, an analysis is made by using R_{NN} values integral-averaged over this band wavelength region.

2.6 Procedure for surface diagnosis

The above algorithm for surface diagnosis is proposed for the most general application. On the other hand, we apply the algorithm to a specified case

of $m=0$ and $\sigma_{II}=0$. That is, the value of β in Eq.(13) is assumed to be constant over the 8 wavelength points which are located near to each other (Section 3.3), and the film-substrate interface II is assumed to be kept smooth. As to the thickness d and σ_I , it is assumed to be $\lambda/(4n_1) > d > \sigma_I$ in the initial stage, where n_1 ($n_1 \cong 2$) is the index of refraction of the film material and $\lambda/(4n_1)$ corresponds approximately to the film thickness of the 1st interference minimum. In the initial stage, there can be a surface film far thinner than the wavelength λ of radiation. In the present diagnosis thickness d is determined first, and then roughness σ_I is determined.

In the film system, with an increase of film thickness d reflectance R at each wavelength oscillates with time t due to the film interference. The 1st order valley and hill, the 2nd order valley and hill, ..., appear successively. In the simple SF model, Eq.(29) results in,

$$R_{NN} = R_{NN}(d) \quad (30)$$

and the film thickness d_{SF} of the minimum and maximum R_{NN} or that of R_{NN} satisfying a differential equation of $\partial R_{NN}(d)/\partial d = 0$ is easily calculated. That is, the film thickness d_{SF} at time t when R_{NN} takes the minimum or maximum values can be known. On the other hand, in the RF model the film thickness d_{RF} depends also on the roughness σ_I of the film. But the dependence is weak, since the average thickness d , which is larger than σ_I , is the dominant factor of phase modification of film interference. If d_{RF} is approximated to be equal to d_{SF} within an acceptable precision, then the average thickness d can be estimated by $d = d_{RF} = d_{SF}$.

Figure 4 examines the relationship of relative deviation $(d_{SF} - d_{RF})/d_{RF}$ in estimated thickness to relative roughness (σ/λ) , for the interference minima and maxima of up to the 3rd order of interference, and for the cases of $\sigma_I = \sigma$ and $\sigma_{II} = 0$. The calculation is made on $\lambda = 0.9, 1.5, 3.5$ and $8.0 \mu\text{m}$ and various σ . The figure suggests that if an error of $\pm 5\%$ is allowed in

Fig. 4 Difference of average film thicknesses estimated by RF model and SF model

the estimation of d , then the following procedure is possible for the d diagnosis in the case of $(\sigma/\lambda) < 0.1$. That is, the minima and maxima are found in the measured time oscillation of R_{NN} . The minima and maxima have corresponding thickness d_{SF} which can be calculated in the SF model. Although this thickness d_{SF} is not equal to the thickness d_{RF} in the RF model for the rough surface, we assume that the film thickness is equal to d_{SF} , which considering an inaccuracy within $\pm 5\%$. Since a number of wavelength points of λ_i distribute at every $(\Delta\lambda/\lambda) = 0.03$ over the spectral region of $\lambda = 0.75 \sim 10.9 \mu\text{m}$, such thickness diagnosis can be made at many points of time t . Resolution of the thickness Δd is of an order of $(\Delta d/d) = 0.02$.

At time t when the average film thickness d is known and $\sigma_{II} = 0$, Eq.(29) turns to,

$$R_{NN} = R_{NN}(\sigma_I) \quad (31)$$

At the time t , there are a number of measured values of R_{NN} at wavelengths rather than the wavelength of the minimum/maximum R_{NN} at the time. Thus, roughness σ_I is determined at the time by a least-squares technique so that the measured values of R_{NN} at the many wavelengths can best fit the calculated values. The order of diffuse reflection by the surface system with rough interfaces can be evaluated by the reflectance ratio β determined in Section 2.4.

3. Experimental Examination of Proposed Surface Diagnosis

3.1 Experimental procedure

We make an experimental examination for demonstrating the performance of the proposed algorithm. An optically smooth surface of nickel of 99.99% in purity is prepared. It is heated from the back in air. The surface is heated from room temperature up to 1100 K with a heating rate of 1 K/s, and kept at the temperature for about 1 h. The surface temperature T^{tc} is measured for reference by a K-type thermocouple of 100 μm in diameter welded on the specimen surface. Emission energy and reflection energy of the surface in this process are measured at 75 wavelength points in the wavelength region of $\lambda = 0.75 \sim 10.9 \mu\text{m}$. The spectrophotometer system and experimental procedure in this experiment are the same as those for the previous work⁽¹⁾. The light sources in the reflection measurement are a tungsten-halogen lamp and a silicon nitride radiation source. The measurement angle for both emission and reflection is $\theta_0 = 15^\circ$. In the analysis of the measured quantities for the surface diagnosis, this value of $\theta_0 = 15^\circ$ is used.

3.2 Results of experiment

Figure 5 shows raw data of surface temperature T^{tc} , emission energy u and reflection energy v mea-

sured at time t . With an increase of time t , the film thickness and roughness of the surface film increase. Many curves for u and v in the figure correspond to the wavelength points of the measurement. In the figure for emission, the shortest and longest wavelengths are $\lambda=1.12$ and $10.9\text{ }\mu\text{m}$ respectively, and in the figure for reflection, the shortest and longest wavelengths are $\lambda=0.75$ and $10.9\text{ }\mu\text{m}$ respectively. Figure 6 shows the data of Fig. 5 in forms of emittance $\varepsilon_N^{\text{tc}}$ and reflectance R_{NN} . Emittance $\varepsilon_N^{\text{tc}}$ is calculated for reference by using the temperature T^{tc} measured by the thermocouple and it is not the result of the analysis. In these figures, clear oscillation of film interference is observed in the curves for the emission as well as in those for the reflection. It is interesting that such clear interference occurs in the emission by the thermal radiation sources^{(1),(8),(10)}, and

that the interference in emission and that in reflection occur simultaneously and complementarily.

3.3 Analysis of experimental results

Experimental results of u and v are analyzed by the algorithm proposed in Section 2. In the diagnosis of the temperature T , and spectra of ε_N , R_{NN} and β of the surface system (Section 2.4), 16 values of u and v at 8 wavelength points of $\lambda=1.12\sim 1.45\text{ }\mu\text{m}$ where the S/N ratio of the measured emission data are best are adopted at each time t . The value of β is assumed to be constant over the 8 wavelength points at each time t . In the diagnosis for the average thickness d and the roughness σ_1 of the film (Section 2.5), 75 values of v at the 75 wavelength points of $\lambda=0.75\sim 10.9\text{ }\mu\text{m}$ are adopted at each time t . The spectrum of optical constant $\hat{n}_1$ of the film has been obtained by analyzing the results of a pre-experiment on radiation interfer-

Fig. 5 Measured quantities: temperature T^{tc} , emission energy u and reflection energy v

Fig. 6 Emittance ε_N^{tc} evaluated by T^{tc} , and measured reflectance R_{NN}

ence in an air-oxidation process of optically smooth surface of nickel. The spectrum is assumed not to depend on temperature T and thickness d . The spectrum of optical constant $\hat{n}_2$ of the substrate nickel is calculated by a function $\hat{n}_2(\lambda, T)$ in Ref.(11) with using the temperature T^{opt} determined in the diagnosis in Section 2.4.

Figure 7 shows the transition of the quantities estimated in the analysis of the present diagnosis: surface temperature T^{opt} , spectrum of emittance ε_N^{opt} , reflectance ratio β , average thickness d and rms roughness σ_t of the surface film. Agreement of temperature T^{opt} and T^{tc} is excellent. The deviation of T^{opt} from T^{tc} is no less than 10 K when enough emission energy is measured at $T > 1000$ K. In particular, at $t > 900$ s when the radiation interference behavior in u and v is weakened, the deviation is

within ± 1 K. It is the result of the present technique of temperature diagnosis, which deal with the spectra of partially diffuse reflection as well as those of emission. Temperature T_{1-8} in the figure is determined by a traditional two-color pyrometry technique⁽¹²⁾ with using emission data measured at $\lambda = 1.12$ and $1.45 \mu\text{m}$. The result of T_{1-8} is not enough on a qualitative behavior level.

Estimated thickness d of the film ranges over $d = 0.08 \sim 3.8 \mu\text{m}$, which is based on the width of the wavelength region covered by the present hardware. The resolution of the thickness d is of an order of $(\Delta d/d) = 0.02$. Considering the R_{NN} - d relationship in the initial stage of the film growth, it can be suggested that change of thickness d of 1 nm is resolved provided the precision of R_{NN} measurement at the shortest wavelength points is better than 1%. The rough-

Fig. 7 Estimated quantities in the present diagnosis: temperature T^{opt} , emittance $\varepsilon_N^{\text{opt}}$, reflectance ratio β , thickness d and roughness σ_1

ness σ_1 of the surface film was estimated in a range of $\sigma_1=0.01\sim 0.71\ \mu\text{m}$. The roughness σ_1 was of an order of 5~10% of the thickness d . It could have been determined on the condition of $(\sigma/\lambda)<0.1$ (Section 2.6).

The thickness d and roughness σ_1 were measured more directly by microscope techniques on a specimen at room temperature cooled after the experimental examination. The results were $d=3.8\ \mu\text{m}$ and $\sigma_1=0.70\ \mu\text{m}$. These values are compared with the estimated values of $d=3.8\ \mu\text{m}$ and $\sigma_1=0.71\ \mu\text{m}$ at the last stage of the experiment at $t=3800\ \text{s}$.

4. Summary

We have proposed an algorithm of thermal radiation spectroscopy for diagnosing temperature and microstructure of real surfaces. The algorithm analyzes near-infrared through infrared spectra of emission and reflection measured simultaneously. The measurement is that in the near-normal direction to the surfaces, which needs the simplest optical setup.

In order to verify the performance of the proposed algorithm, an experimental examination was made on a metal surface in a high-temperature air-oxidation process. Emission spectra in a wavelength region of $\lambda=1.12\sim 10.9\ \mu\text{m}$ and reflection spectra in a region of $\lambda=0.75\sim 10.9\ \mu\text{m}$ were measured at every 2 s and analyzed to estimate the transition of the surface temperature T^{opt} , spectra of emittance $\varepsilon_N^{\text{opt}}$ and reflectance R_{NN} of the surface, reflectance ratio β of the surface, average thickness d and rms roughness σ_1 of the surface film. As results of this examination, the surface temperature of an order of 1100 K was estimated within an inaccuracy of 10 K. The thickness d of the film was estimated in a range of $d=0.08\sim 3.8\ \mu\text{m}$ with a precision of $(\Delta d/d)=0.02$. The roughness σ_1 was estimated in a range of $\sigma_1=0.01\sim 0.71\ \mu\text{m}$. Reality of a new non-contact in-process diagnosis for temperature and microstructure of real surfaces has been demonstrated.

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