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Original Research Paper

DEVELOPMENT OF CLEANING EVALUATION APPLICATION USING HUE CHANGE AS A PARAMETER

TORU TSUKIZAWA, TATSUYA NAKAMURA, MASARU OYA✉
Graduate School of Environment and Information Sciences, Yokohama National University 79-7 Tokiwadai, Hodogaya-ku, Yokohama, 240-8501, Japan.
e-mail: mooon0128@yahoo.co.jp, kotatsu-97b@ezweb.ne.jp, moya@ynu.ac.jp

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ABSTRACT

In the cleaning field, there is a method to evaluate the amount of soiling by processing the color signal of a soiled sample's image using the Kubelka-Munka formula, but this method could not be adapted to the case where the hue changes drastically during cleaning or washing. In this study, we aimed to construct a system to quantify the amount of soiling corresponding to the hue change. In other word, this is the sturdy for image analysis of individual measuring devices, which is an extension of the cleaning rate analysis, especially in cleaning tests. We conducted a cleaning experiment using soiled fabrics of iron oxide as a representative dirt model of drastic hue changes because its hue changes from yellowish-brown to black by reduction process. Then we calculated the cleaning rate of the dirt on the soiled fabric by the image analysis, conventional method using Kubelka-Munka formula, and X-ray Fluorescence (XRF) analysis, and we evaluated the accuracy of the proposed method of the image analysis by comparing the results of XRF. As a result, first it was found that the hue changes of iron oxides due to reduction could be clearly captured as a color signal, and the degree of reduction of iron oxides could be determined from the Chromaticity Diagram Value, xy value. Using the results, we were able to construct an application that can easily determine the amount of soiling adhesion by recalculating the hue change as a variable. The cleaning rate calculated by the image analysis application showed a significant improvement compared to the one calculated by the conventional appearance evaluation method. This achievement has greatly expanded the range of objects for which the amount of soiling from the exterior can be evaluated.

Keywords: cleaning, color, hue, image analysis, iron dioxide, Kubelka-Munk formula.

INTRODUCTION

Detergency is generally evaluated from the change in the amount of dirt before and after cleaning. One of the most accurate methods is the extraction-chemical quantification method. However, it has the disadvantage of requiring a lot of time and effort for extraction, reaction processing, and chemical analysis. Therefore, in many actual cleaning sites, a method that evaluates the degree of dirt adhesion based on appearance instead of extraction-chemical quantification is adopted. One of the methods used to quantitatively evaluate the degree of soiling is the K/S value derived from the Kubelka-Munka formula. This value represents the ratio of light attenuation due to absorption and scattering from reflectance. The advantage of this quantification is a simple and non-destructive way of estimating the amount of soiling. As the amount of dirt decreases, the color density decreases, and the reflectance value increases, and on the other hand, as the amount of dirt increases, the color density increases, and the reflectance value decreases. Based on the color density about this, we calculate the parameters that are theoretically proportional to the amount of dirt. This method has been used for many material quantifications, for example in the following cases: varnish layers applied to paintings (Kirchner *et al.*, 2018), carbon black on fibers (Gotoh *et al.*, 2015; Gotoh *et al.*, 2016) Virgin olive oil content (Gomez-Robledo *et al.*, 2008), the concentration of cellulose degradation products (Micheli *et al.*, 2018), small amounts of contaminants such as organic fractions of aerosols (Ghauch *et al.*, 2006).

However, some combinations of dirt types and cleaning process methods are excluded from the above principles because the hue of the substance changes drastically before and after the cleaning process. For example, due to the effect of a reducing agent practically used to remove iron rust dirt, iron rust that exists in yellowish-brown or red after treatment may be reduced to black ferric tetroxide (Ohtsuka *et al.*, 2015; Suzuki *et al.*, 2008). In addition, curcumin contained in curry pigments changes from yellow to red under alkaline conditions (Suksulap *et al*., 2014), which is caused by depurtonation of curcumin (Chen *et al*., 2021). Under such conditions, the K/S value calculated by Kubelka-Munka formula can't be applied and the quantification using surface dye concentration would become difficult. This is because the relationship between the surface dye concentration and the amount of soiling is broken. The conditions that quantification from surface dye concentration is possible are, as mentioned above, when the amount of stain increases as the surface reflectance decreases, and in such cases substances such as iron oxides have been accurately quantified using the kubelka-munka equation (Goncalves *et al*.,2012; Milhomem *et al*., 2013).

In this study, we propose a simple method by image analysis for quantifying dirt substances from the external appearance, even when the hue changes drastically before and after processing. It is an image analysis of individual measuring devices, especially aiming to be used for cleaning rate analysis in cleaning tests.

In the cleaning field, as methods to evaluate the amount of dirt from the appearance, a method to quantify the amount of soiling based on the image data obtained from a digital camera has been developed in addition to the conventional method using a spectrophotometer. For example, RGB values were extracted using image data taken by a digital camera (Canon Digital SLR Camera EOS 7D) instead of using the spectrophotometer (Miyazaki *et al*., 2003). In addition, a method to calculate XYZ values using multiple regression analysis from the Macbeth color checker was examined (Ishikawa *et al*., 2005). Using them, there are also studies on quantification of dirt on metallic luster (Muto *et al*., 2011), colored fabric (Kojima *et al*., 2015a), model oil-based stains (Ishikawa *et al*., 2006), and pigments or model particle stains like iron oxide particles (Ishikawa Y *et al*., 2008). Image analysis has also been applied to evaluate the uniformity of soiled fabric (Kojima *et al*., 2015, b) and to quantify colorless non-polar oily stains from UV images (Keiko Sugita *et al*., 2021). The changes in hue and saturation can be shown as numerical values by converting the RGB values of the target measurement object to the X, Y, and Z values of the XYZ color system and obtaining the color mixing ratio Chromaticity Diagram Value, xy value, (Dobrusina *et al*., 2011) and the relationship between the xy value change and the amount of substance will be applied to quantify the unknown substance to be measured.

In this study, iron rust dirt is used as a dirt model because it changes its color from yellowish-brown to black after being cleaned with a reducing agent. We create an image analysis application that incorporates an algorithm for determining the relationship between the color mixing ratio xy value and the amount of material measured by an analytical instrument. And we develop a simple method to quantify dirt from image signals even when the appearance hue changes drastically.

MATERIAL AND METHODS

IMAGE ANALYSIS APPLICATION

Firstly, the application was created using VISUAL BASIC 2019. The image analysis consists of the calculation step of the tristimulus value XYZ using the multiple regression equation and the calculation step of the ΣK/S value which is the apparent dirt quantity parameter. First of all, in the calculation step of XYZ value, the color image of 24 colors of Macbeth color checker (Gretag Macbeth) is taken by Canon Digital SLR Camera EOS 7D, and the RGB value of each of 24 colors is obtained from the image file. Then, the second-order multiple regression equation between each XYZ value described in the Macbeth color checker is obtained, and the conversion equation from RGB values to XYZ values is obtained. At this time, the XYZ value described in the Macbeth color checker is used as an objective variable and each RGB value is used as an explanatory variable. 3 regression equations are obtained in total for each of the X, Y, and Z values, and the conversion equation for the Y value is shown below as a representative. (Eq.1).

$$\mathrm{Y} = \mathrm{a} \times \mathrm{R} + \mathrm{b} \times \mathrm{G} + \mathrm{c} \times \mathrm{B} + \mathrm{d} \times \mathrm{RG} + \mathrm{e} \times \mathrm{GB} + \mathrm{f} \times \mathrm{BR} + \mathrm{g} \times \mathrm{R2} + \mathrm{h} \times \mathrm{G2} + \mathrm{i} \times \mathrm{B2} + \mathrm{j} \qquad (1)$$

Y: lightness, R: red component of color information, G: green component of color information, B: blue component of color information, a〜i: each coefficient calculated by the multiple regression equation, respectively.

Applying Eq. 1 to each of the 24 colors yields the determinant (Eq. 2), and from Eq. 3, it is possible to find the determinant A, which is each constant from a to i (Eq. 4).

$$\mathrm{Y} = \mathrm{CA} \qquad (2)$$

$$\mathrm{CTY} = \mathrm{CTCA} \qquad (3)$$

$$\mathrm{A} = \mathrm{CTY} \qquad (4)$$

Then C, A, and Y are the following determinants, respectively.

$$C = \begin{bmatrix} R_1 & G_1 & B_1 & RB_1 & GB_1 & BR_1 & R_1^2 & G_1^2 & B_1^2 & 1 \\ R_2 & G_2 & B_2 & RB_2 & GB_2 & BR_2 & R_2^2 & G_2^2 & B_2^2 & 1 \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \\ R_{24} & G_{24} & B_{24} & RB_{24} & GB_{24} & BR_{24} & R_{24}^2 & G_{24}^2 & B_{24}^2 & 1 \end{bmatrix} \tag{5}$$

$$\mathrm{A} = \begin{bmatrix} a \\ b \\ \vdots \\ i \end{bmatrix} \tag{6}$$

$$Y = \begin{bmatrix} Y_1 \\ Y_2 \\ \vdots \\ Y_{24} \end{bmatrix} \tag{7}$$

The following X and Z values are calculated in the same way.

The formula for converting the XYZ values obtained from the above regression equation to xy values representing lightness and hue is as follows.

$$x = \frac{X}{X+Y+Z} \tag{8}$$

$$y = \frac{Y}{X+Y+Z} \tag{9}$$

In the step of calculating the ΣK/S value, which is an apparent dirt quantity parameter, the XYZ value is obtained from the RGB value of each pixel of the dirt image data using the multiple regression equation (Eq. 1), the Y value (brightness) is converted from a percentage to a decimal point, and the K/S value is obtained from the Kubelka-Munk formula (Eq. 10) as the surface reflectance obtained. The ΣK/S value, which is the sum of the K/S values of each pixel, is calculated as an index proportional to the degree of contamination.

$$\frac{K}{S} = \frac{(1 - 0.01Y)^2}{2 \times 0.01Y} \tag{10}$$

K: Light absorption coefficient, S: Light scattering coefficient

Next, we will compare the accuracy of the values obtained by the above procedure with the values measured by the spectral color difference system. Y, x, and y values of four-color fabric (Fig. 1) are measured using an image application and two spectral color difference systems. Three sheets of each color were prepared, and each sheet was measured twice in total, front and back. In other words, the average value of six measurements per color was obtained, and that was used as the measurement value of each measuring instrument.

Fig. 1. *Colored Fabric (Blue, Green, Red, White).*

In addition, the blur of the measured value due to the change of the shooting environment was also considered. Even for the same measurement object, the color information of the image often differs due to changes in illuminance, variations inherent in the electric light, and errors in the color reproduction of the camera (Chen *et al.*, 2020). Therefore, we will also check the accuracy in such cases. As a measurement method, the red fabric shown in Fig. 1 was photographed for 10 days to check how much the color values (X, Y, Z, x, y) could change.

MATERIALS TO PREPARE THE ARTIFICIALLY SOILED FABRIC MODEL

The materials used to prepare the artificially soiled fabric model were cotton canequim（Laundry Science Association in Japan, Sodium Hydroxide (FUJIFILM Wako Pure Chemical Co.), Iron(III) Chloride Hexahydrate (FUJIFILM Wako Pure Chemical Co.). And Hydroxylammonium Chloride(Wako Pure Chemical Industries, Ltd), 1,10-phenanthroline monohydrate (Junsei Chemical Co, Ltd.), and Sodium Acetate (FUJIFILM Wako Pure Chemical Co.) were chosen for quantitative analysis of iron ions, while Oxalic Acid (Wako Pure Chemical Industries, Ltd), Ascorbic Acid (Wako Pure Chemical Industries, Ltd), Hydrochloric Acid (FUJIFILM Wako Pure Chemical Co.), Phosphoric Acid (Wako Pure Chemical Industries, Ltd), DL-Malic acid (Wako Pure Chemical Industries, Ltd), Citric Acid (FUJIFILM Wako Pure Chemical Co.), and Acetic Acid (FUJIFILM Wako Pure Chemical Co.) were used as acid agents in the cleaning experiments.

CREATION OF IRON OXIDE MODEL

Canequim white fabric cut into 5cm x 5cm is dipped into 20wt% iron chloride solution. The fabric is then squeezed with a mangle to adhere evenly, reacted with 1wt% NaOH boiling solution, and dried. The created contamination model is qualitatively analyzed by X ray diffraction (XRD) and scanning electron microscopy (SEM) to confirm the reproducibility of the iron rust model. In addition, immersion cleaning with six kinds of acids is performed to confirm the same cleaning behavior as that of conventional iron rust.

RELATIONSHIP BETWEEN XRF AND PHENANTHROLINE ABSORPTIOMETRY

In this study, XRF was used to measure the iron content of the iron rust model, which can be analyzed non-destructively and simply. The reliability of the iron content determined by this XRF will be compared with that determined by the phenanthroline absorption method. Phenanthroline absorption spectrophotometry is a technique used for the determination of iron content (Pradhan *et al*., 2019; Hayati *et al*., 2014). This colorimetric determi nation is done by using the complexation of three molecules of 1,10-phenanthroline (ortho-phenanthroline) with ferrous ions (Fe^{2+}) (Kozak *et al*., 2011). Therefore, as a method to compare the iron content measurement by XRF and phenanthroline absorbance method, firstly, 13 samples of the fabricated iron rust model will be immersion washed with 0.5% oxalic acid at 40°C. Since the variable range of iron content in a normal cleaning experiment is about 0% to 9%, the iron content was adjusted so that the range was included between 0% and 11%. Their iron contents were measured by XRF, and as for the phenanthroline absorbance method, it was done as follows: After all the iron ions were dissolved by immersing the iron rust model in 2.5% HCl solution, 2.0 ml was squeezed out and 1.0 ml of 0.3 M hydroxylamine hydrochloride solution was added. After the reaction is complete, 5.0 ml of 1.0 M sodium acetate solution and 5.0 ml of 0.25% 1,10-phenanthroline solution are added to the flasks and mixed by inverting. Deionized water is added to make the total 50 ml and this is measured at 510 nm absorption wavelength to obtain the absorbance. The measured values of the above two points are compared and their effectiveness is examined.

CLEANING TEST AND CALCULATION OF CLEANING EFFICIENCY

First of all, the relationship between the hue and the amount of dirt of the iron rust model made by the above method is investigated: 50 ml of 0.5 wt% oxalic acids is immersed in a solution at 40°C for 5 minutes each, and the xy value is measured from the application for the hue, and the mass percent of iron (Fe wt%) is measured for the amount of dirt using an X-ray fluorescence spectrometer. Similarly, the xy value and Fewt% are measured when 50 ml of 0.5 wt% oxalic acid is immersed in a solution at 40°C for 5 minutes at a time, and the correlation is confirmed. After confirming the correlation between the hue and the amount of iron from the relationship between x-Fewt% and y-Fewt% for two kinds of iron rusts, the amount of iron is estimated from the hue using the approximation curve. Then, the cleaning experiment to confirm the accuracy of the above approximation curve is described below.

Similarly, the iron rust model created by the above method is subjected to immersion cleaning with ascorbic acid. At that time, the cleaning conditions are performed by changing the three parameters of temperature, concentration, and cleaning time, and the cleaning rate is calculated by the following three methods (from the amount of change in the mass percent of iron content using XRF, from ΣK/S using image application, and from the calibration curve of hue and iron content). The goal is to obtain agreement between the results of the washing rate determined by quantification and the results of the washing rate determined from the calibration curve under all conditions.

RESULTS

IMAGE ANALYSIS APPLICATION

First of all, the determination coefficients of all three conversion formulas from RGB values to XYZ values obtained from the application created by the method described above were all 0.95 or higher, which is a good result (Table 1). Therefore, it was confirmed that X, Y, Z, x, and y values could be calculated with high accuracy for various colors. In addition, the values of the spectrophotometers and the values calculated by the image application are generally consistent (Fig. 2). In addition, the coefficient of determination of the average value of the spectrophotometer and the average value of the image application was calculated to be Y:0.997 x:0.998 y:0.990, suggesting the calculation of five types of color information values for various colors with high accuracy. Finally, from Fig. 3 and Table 2, it was confirmed that there was no change in color values with the passage of days and that the values were similar to those of the spectroscopic colorimeter, suggesting that the system was able to cope with a changing photographic environment.

Table 1. *R^2 value and standard error of regression equation of X, Y, and Z*

	R^2	Standard error
X	0.9964	5.012
Y	0.9888	4.572
Z	0.9586	9.514

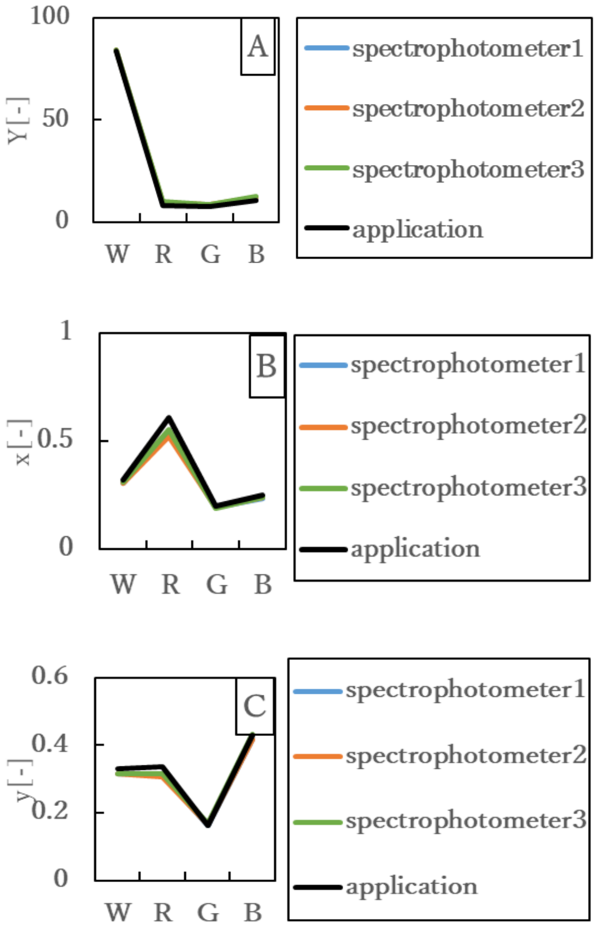

Fig. 2. *4 colors (White, Red, Green, and Blue) vs Y(A), x(B), y(C).*

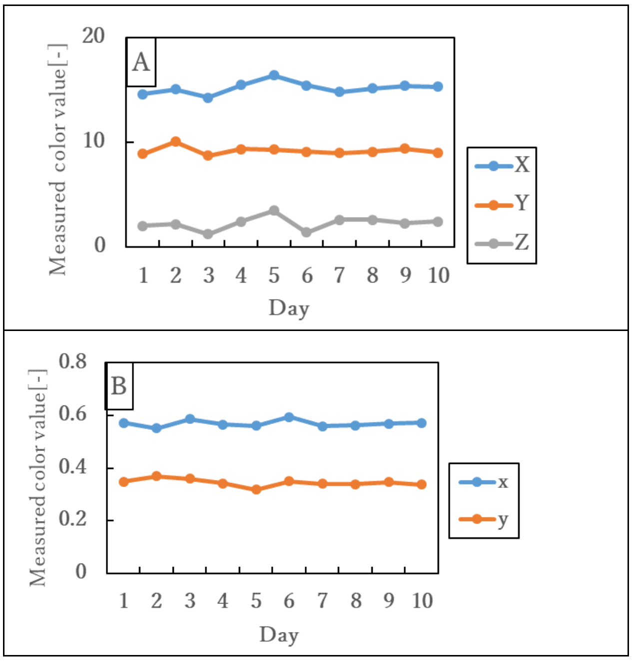

Fig. 3. *Change in the measured X, Y, and Z values (A) and x and y values (B) with the passage of days.*

Table 2. *Color value of red fabric measured by application and spectrophotometer*

	Average value	**Standard error**	**Value of spectrophotometer**
X	15.76	0.6175	17.78
Y	9.188	0.3816	10.24
Z	2.287	0.6716	4.807
x	0.5696	0.01359	0.5415
y	0.3454	0.01414	0.312

CREATION OF RUST DIRT MODEL

The results of the qualitative analysis of the iron rust model (Fig. 4) created in this section will be described.

From Fig. 5, the peaks around 27 °, 35 °, 39°, 57 °, 62 °, which are specific to β-FeOOH, are confirmed in XRD (Reagan *et al*., 2016). The sample used in this study was prepared from a glass slide as a substrate for the measurement. This is to avoid detecting the peak of the substrate. The hollow crystal structure characteristic of β-FeOOH was confirmed from Fig. 6 (An *et al*., 2017). It is said that β-FeOOH is produced in the presence of chlorine. Therefore, iron oxide dirt adhering to the inside of water pipes is also β-FeOOH. When the particles in the inside of the water pipe in a 45-year-old building in Nerima Ward were checked by SEM, crystals similar to those in the iron rust model were observed. The results of the cleaning experiments using various acids showed that hydrochloric acid, oxalic acid, and phosphoric acid, which are considered to be effective against iron oxides, showed high cleaning rates, while other acids showed low cleaning rates (Fig.7). The immersion washing conditions were as follows: acid concentration: 1 wt%, washing temperature: 60 °C, washing time: 60 min. In other words, this showed the same cleaning behavior as that of conventional iron oxides.

From the above, it was confirmed that the iron rust model created was similar to β-FeOOH in the water pipe, and it was possible to replicate the iron rust dirt that is generated in daily life.

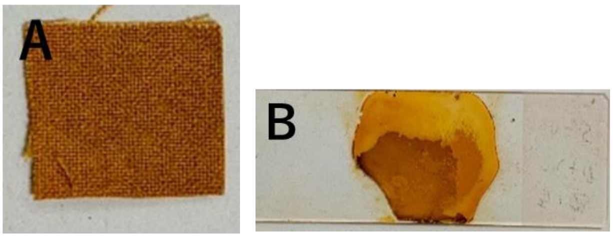

Fig. 4. *The picture of the Iron oxide model on fabric (A) and the microscope slide (B).*

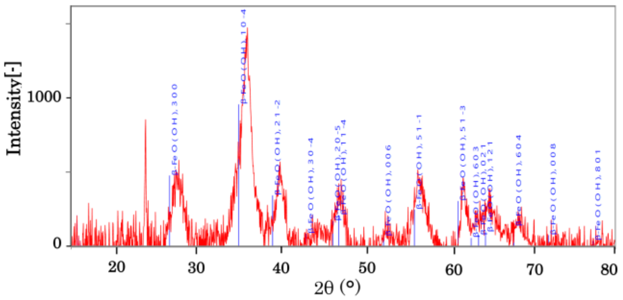

Fig. 5. *XRD results of iron oxide model.*

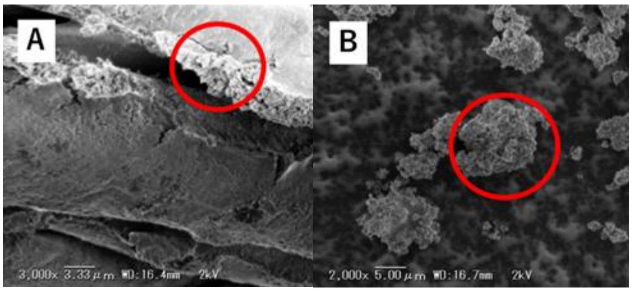

Fig. 6. *Observation results of iron oxide model (A) and water pipe internal dirt (B) by SEM.*

RELATIONSHIP BETWEEN XRF AND PHENANTHROLINE ABSORPTIOMETRY

From Fig. 8, the coefficient of determination of the calibration curve for only the plots with iron content up to 9.26 % is 0.987, which is a good result. The coefficient of determination is 0.888 when plots with iron content of 10.14 % and 11.07 % are included. In other words, the calibration curve can be defined as a first-order approximation when the iron content is up to 9.26 %, and it is possible to regard the value measured by the X-ray fluorescence spectrometer as the iron content.

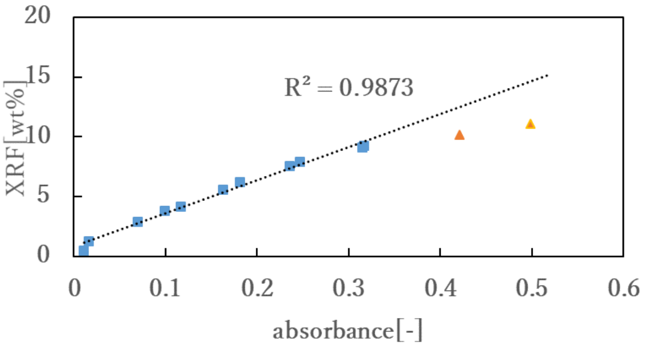

Fig. 8. *Relation between absorbance and XRF*

CLEANING TEST AND CALCULATION OF CLEANING EFFICIENCY

Next, the correlation between iron content and hue will be mentioned. In the removal of iron rust on industrial surfaces, immersion removal using acid agents is commonly used (Daurelio *et al.*, 1997; Lee *et al.*, 2007). Among them, oxalic acid is widely used (Tzagkaroulakis *et al.*, 2017) and is expected to provide a high cleaning rate at low concentrations.

The color values (x,y) of the samples were measured by oxalic acid and ascorbic acid (Fig. 9). Fig.10 shows that there is a strong correlation between the amount of iron and the hue of the yellowish-brown iron rust soiled model and the blackened ascorbic acid model (Fig.11). It is suggested that there is a correlation between the amount of iron and the concentration of iron because the adhesion of the particles to the fiber is the same in the case of blackening as in the normal case. Therefore, from these two calibration curves, the amount of iron was measured from the measured value of the hue.

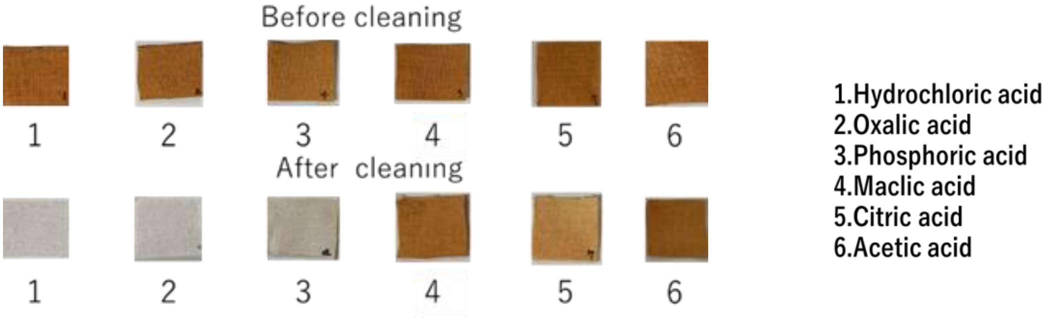

Fig. 7. *Cleaning results by using 6 kinds of acids: 1. Hydrochloric acid, 2. Oxalic acid, 3. Phosphoric acid, 4. Malic acid, 5. Citric acid, 6. Acetic acid.*

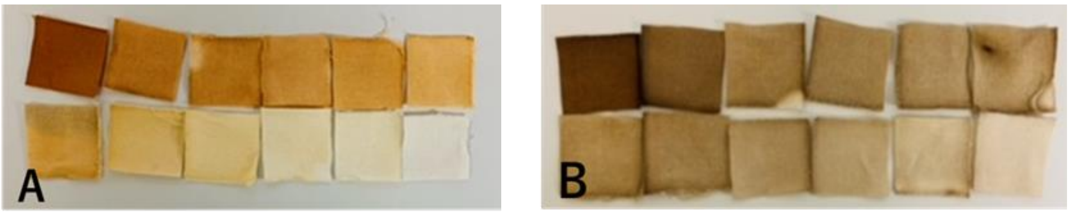

Fig. 9. *The State of soiled fabric after cleaning test with oxalic acid (A) and ascorbic acid (B).*

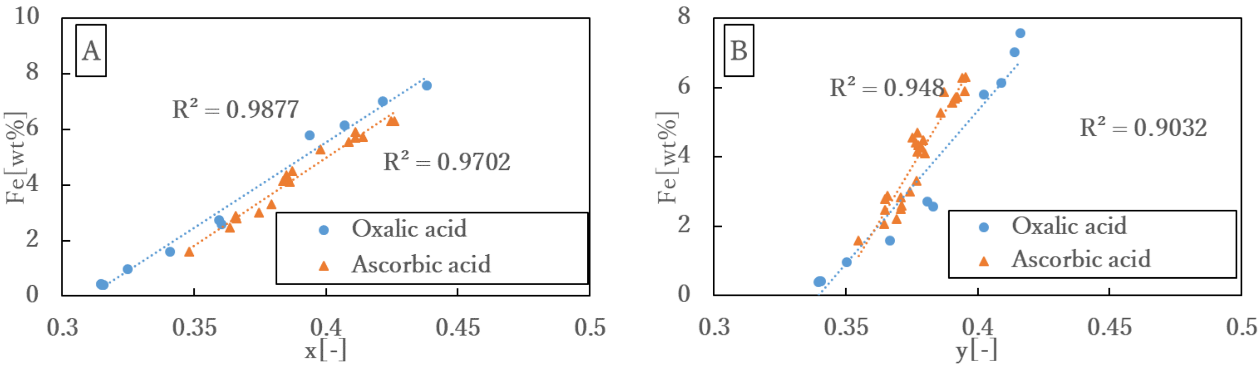

Fig. 10. *The iron amount measured by XRF and x value (A), y value (B).*

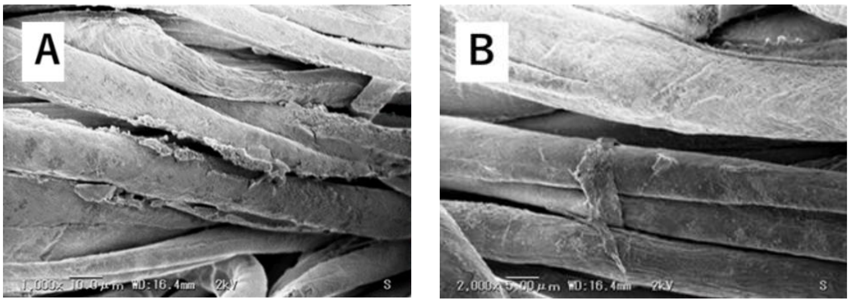

Fig. 11. *SEM observation of the state of Iron rust model (A) and after immersion in ascorbic acid (B).*

Finally, the results of immersion experiments in ascorbic acid are described. Here, the washing experiment was carried out using one of the following variables: washing time, concentration, and temperature (Fig. 12). In A, washing time was used as a variable, in B, concentration was used as a variable, and in C, the temperature was used as a variable. From the three graphs, the cleaning rate calculated by using the calibration curve above was close to the cleaning rate by XRF. The cleaning rate by K/S value, which is used for quantitative evaluation from appearance, shows negative values in some places. The cleaning rate calculated by the K/S value, which is conventionally used as a quantitative evaluation method from the appearance, was much improved.

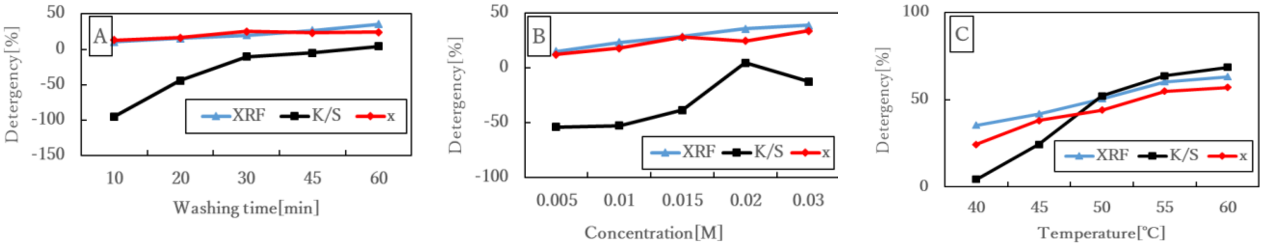

Fig. 12. *Detergency of iron oxide model when cleaning time (A), concentration (B), and temperature (C) discharged.*

DISCUSSION

In this study, we proposed a method to quantify the substance even when its hue changed to black after treatment. In the results of immersion cleaning with ascorbic acid using iron oxides as a representative model, the cleaning rate calculated from the relationship between the change in hue and the change in iron content and the cleaning rate calculated by quantifying the iron content generally agreed with each other. In the case of the conventional method using the K/S value, the amount of dirt adhesion increased when the model turned black after washing. In this study, only iron oxide dirt was examined, but the method can be applied to other materials by considering the materials before and after treatment independently and comparing the hue change and the amount of material change.

When conducting this study, we attached iron oxide dirt to a white fabric to create a model. Adhering the particles to the white fabric had the advantage that the surface area was large and it was easy to see the changes during cleaning. Compared to the case where the particles were attached to a surface such as a glass slide, it was possible to clean them with a low concentration of acid. It is thought to be effective as a contamination model at the laboratory level when searching for a removal method for iron oxides, which has not yet been established. However, due to the influence of gaps between fibers and shadows, the color values produced by the image application were lower than those measured by the spectral color difference (Abril *et al*., 2008). Therefore, it is suggested that the reliability of the absolute color values is lower than that of the color difference system. In this study, XRF was used instead of the extraction method to determine the iron content, because it is non-destructive and simple to determine the iron content (Jacobi *et al*. ,2007; Chen *et al*. ,2019; Mejia-Pina *et al*. ,2016; Ene *et al*. ,2010; Horf *et al*., 2021). Comparison of the iron content values measured by XRF with the results of the extraction method showed that the iron content could be accurately determined up to the fiber increases as the iron content increases. The iron particles near the fiber are not measured, and only the outer surface is measured, so it is thought that the measurement results are smaller than the actual amount of iron attached. Therefore, it was shown that XRF can be used for simple determination if the amount of material to be measured is within the measurable range.

The results of the cleaning experiments are discussed in the last section. In the conventional appearance evaluation, the cleaning rate became -, that is, the amount of contamination increased after cleaning. As shown in the XRF results, the iron ions are dissolved by immersion in ascorbic acid, but the hue on the surface becomes darker, and the surface reflectance, which is the Y value, decreases significantly. As a result, the amount of apparent dirt increases after washing, and the washing rate is considered to have a negative value. In the case of immersion cleaning at a temperature of 50°C or higher for 60 minutes, the cleaning rate was the same as the XRF value (Fig.12:C), but this was because the blackened iron rust was dissolved by the passage of time and the blackened part itself was considerably reduced. This is because the blackened iron rust was dissolved by the passage of time and the blackened part itself was decreased. The difference between the cleaning rate calculated by the calibration curve and that by XRF gradually increased as the cleaning time increased. This may be due to the dissolution of particles inside the fiber that cannot be judged on the surface. In such a case, the detergent ratio calculated from the calibration curve would be less than the detergent ratio calculated from the actual iron content. Therefore, depending on the substrate and treatment conditions, there may be some discrepancies with the actual values. However, as a simple quantitative method, it is expected to improve significantly compared with the conventional appearance method.

As a future prospect, we aim to establish the reliability of this method further by examining materials other than iron oxides whose hue changes drastically after treatment. In addition, we will investigate the error of the color value measurement when the image taken is not corrected by Macbeth Color Checker as in this study, and we will also examine the adaptability to off-site use.

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