

Research Article

Novel Chromatic Technique Based on Optical Absorbance in Characterizing Mineral Hydraulic Oil Degradation

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A low cost, compact, real-time, and quick measurement optical device based on the absorbance of white light, which comprised of photodiodes in a 3-element color-sensor, feedback diodes, water and temperature sensing element, and so on, was developed and tested in low absorption mineral oil. The device, a deviation from conventional electrical, mechanical, and electrochemical techniques, uses color ratio (CR) and total contamination index (TCI) parameters based on transmitted light intensity in RGB wavelengths for oil condition monitoring. Test results showed that CR corroborated CIE chromaticity (X - Y Coordinates) and increased with oil degradation unlike Saturation S and Hue H . CR was found to be independent of the particulate contaminants of oil, but dependent on chemical degradation. TCI depended on both chemical degradation and particulate contaminants in oil, being most sensitive in the blue wavelength range and least in the green. Furthermore, results agreed with those of viscometry, total acid number (TAN), and UV-VIS photospectrometry. CR and TCI gave clearer indication of oil degradation than key monitoring parameters like TAN and were found to be effective criteria for characterizing the degradation of hydraulic mineral oils.

1. Introduction

1.1. Background. Oil contamination could be chemical (oxidative and thermal degradation [1], dissolved and emulsified water) or physical (from wear particles, dust, free water, and air). Besides, oil can also degrade by photodegradation (from exposure to the sun ultraviolet rays) [2, 3]. As oils degrade in service, chemical and physical changes inevitably occur. These include change in acidity due to oxidation and additive depletion, viscosity change due to formation of higher molecular weight components, and increased interfacial wear of mating pairs. These changes affect the oil performance.

Oil performance characteristics can be detected by total acidity number (TAN), viscometry, UV-VIS photospectrometry, and so forth. However, these methods mainly measure a single parameter and assume no machinery malfunctions during the measurements. Single parameter sensors only

provide a narrow view of a lubricant quality. Hence, finding a set of measurands that unambiguously identifies the emergence of oil degradation is important. This set of measurements must satisfy statistical criteria so that rational limits can be set on deviations. Subsequently, development of reliable sensor to quantify these measurands becomes imperative. To increase measured data adequacy and reliable estimation of tribocondition, development of online integrated system is strongly desirable.

Recently, various oil monitoring sensors have been proposed. Kumar et al. [4] developed an online optical color sensor which could transform oil darkness during degradation into electrical resistance. Light intensity passed through the oil from one end is recorded at the opposite end. Change in oil opacity derived from degradation is detected as change in light absorbance which is converted to electrical resistance. Similarly, Turner and Austin [5] proposed an

electrical technique for online condition monitoring of lubricating oils, by tracking oils dielectric constants, the ratio of the permittivity of oil to the permittivity of free space, using interleaved-disc capacitors. Khandaker et al. [6] used chromatic modulation technique to detect change in oil condition not by detecting intensity changes at particular wavelengths but by monitoring the sum of the contributions of relative changes at all wavelengths within a spectral power distribution. Electrical inductance techniques based on change in inductance of transducer coils due to the presence of metallic particles had been proposed by Whittington et al. [7] and Flanagan et al. [8]. Such devices are more sensitive to wear particles, compared to oil chemical degradation which usually precedes wear particle generation at tribo-interfaces. Besides, the ≤ 100 micron size particles detected with their devices pose lesser problems to hydraulic oils due to filtration. Other conventional oil sensors such as Delphi sensor [9] and Conti-Temic sensor [10] are based on dielectric constant principle. Lubrigard [11] and Kittiwake [12] based on dielectric/Tan-delta measurement, Flowtonics/Impact sensor [13] based on electrochemical impedance measurement, whereas the Voelker sentinel sensor [14] is based on conductivity measurement of polymeric bead matrix. Another current trend in the art of oil monitoring is the development of miniaturized devices based on optical absorption spectroscopy [15, 16]. Also, Chen et al. [17] used multi-RGB (7-nos RGB) color sensors for the measurement of LED ceiling light in air, not oil. The results of these state-of-the-arts sensors could be masked by the presence of wear particles and polar contaminants in oil. Hence, there is need for a paradigm shift.

In this work, a multiparameter detector based on optical absorption in a 2 cm thick oil-column and integrating 3 sensors to quantify 4 measurands (chemical degradation, total contamination, water content, and temperature) is proposed. With the device, oil chemical degradation was evaluated by color ratio (CR), and total contamination was measured by optical density in RGB wavebands. Water content was measured as relative saturation (%RS) by using air-humidity sensor-element (*HIH-3610*, Honeywell Inc., USA) modified for oleoapplications. Temperature was measured by a temperature sensor-element (*TC1047*, Microchip Inc., USA). Analyses of various oil samples from machines and artificially aged samples were performed, and results compared with other conventional techniques.

1.2. Sensor Measurement Principle. As light particles pass through matter, it is partly absorbed based on the properties of the material. For a radiation model traveling in a low absorption medium of molar absorptivity ϵ with uniform opaque absorption sites of concentration c as in Figure 1, Lambert-Beer's law, which is a means of expressing how light is absorbed by matter could be applied. Lambert-Beer's law states that the amount of light emerging from a sample is diminished by three physical phenomena, namely: (1) the amount or concentration c of absorbing material within the pathlength, (2) the optical path length, and (3) the molar absorptivity ϵ which is the probability that the photon of

that particular wavelength will be absorbed by the material. Mathematically, absorbance A_b becomes:

$$A_b = \ln\left(\frac{I_i}{I_o}\right) = \epsilon cl = k_\lambda l = D, \quad (1)$$

where k_λ is the absorptivity coefficient, I_i and I_o are the inlet and outlet intensities, respectively. There is color change associated with change in absorptivity coefficient ($k_\lambda = f(\epsilon, c)$). As oil degrades in service its k_λ , color, and absorbance also change. It is known that oil color becomes darker with the service time; hence, there is a color shift to long wavelength ranges. Therefore, the ratio of transmitted light intensity in the red (R) to green (G) wavelengths, CR, increases during oil degradation.

For a given spectral sensitivity S' of the detector photoreceiver, the output voltage (U) is proportional to the output light intensity (J), hence, CR becomes

$$CR = \frac{J_R}{J_G} = \frac{U_R}{U_G}. \quad (2)$$

Therefore, the longer an oil service time, the higher its CR. The device also measures a second condition parameter—TCI in RGB wavebands to evaluate the total contamination.

Oil TCI may be estimated by optical density [18, 19], which depends on the content of oxidation and ageing products, dust particles, water, and wear debris. From (1) TCI can be expressed as

$$TCI = D = \ln\left(\frac{I_i}{I_o}\right) = \ln\left(\frac{U_i}{U_o}\right). \quad (3)$$

For the purpose of sensor design implementation in algorithm initialization a baseline of $U_i = U' = 2000$ mV was applied [20]. Hence,

$$TCI^{f,u} = \ln\left(\frac{U'}{U^{f,u}}\right) = \ln\left(\frac{2000}{U^{f,u}}\right) \quad \text{in RGB}, \quad (4)$$

where U' is output voltage with sensor out of fresh (f) and used (u) oil.

2. Experimental Details

2.1. Sensor Probe Description. Figure 2 shows a design of the oil degradation detector probe developed (Kong et al. [19]—USA Patent application number 20080024761). The probe comprises the housing (8) in which oil chemical degradation and total contamination, water content and temperature sensors are mounted. Optical radiation from the white LED (1) in Figure 2 enters into a 2 cm thick oil-column through the optical window (3) and is incident on the RGB color sensor (2) through the optical window (4). Feedback photodiode (5) is used to stabilize the optical power of the emitter. The emitter (1) and color sensor (2) are connected to a preamplifier PCB under cover (11) by electrical wires (10). The entire detector probe is connected by cable (12) to a microcontroller (not shown in Figure 2). Water sensor (6) and temperature sensor (7) are glued to housing (8) by epoxy. Protective mesh (9) is also mounted on the housing (8) to isolate air bubbles and secure optical windows from mechanical damage.

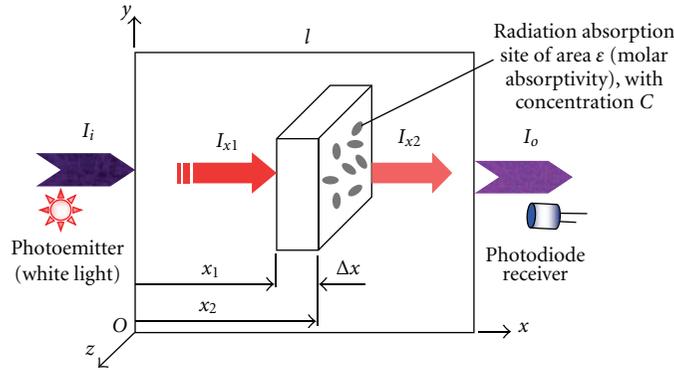


FIGURE 1: Model optical radiation passing through a hypothetical absorption medium.

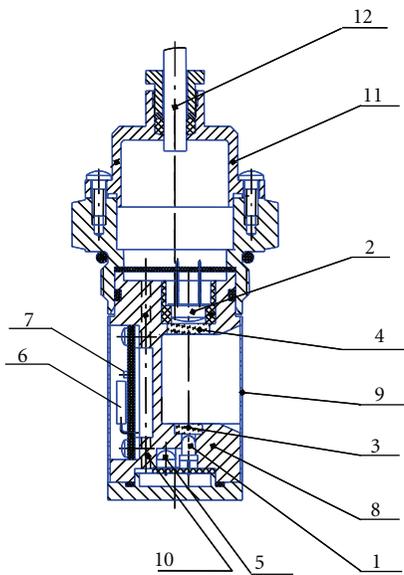


FIGURE 2: Probe for chromatic parameter device ((1): white LED, (2): RGB color sensor, (3, 4): optical window, (5): feedback diode, (6): water sensing element, (7): temperature sensing element, (8): housing, (9): protective mesh, (10): electrical wires, (11): pre-amplifier PCB, (12): connecting cable).

2.2. *Materials and Methods.* Three mineral hydraulic oils, T100, DTE-24 (#1, #2), and DTE25 oils (*Exxon-Mobil*) were obtained from the hydraulic systems of a steel factory (*POSCO, Korea*), were used for calibration and testing. Bulk oil operational temperatures of the hydraulic systems were 20–30°C and 50–65°C in winter and summer, respectively. For these oils, A sample is the fresh oil sample, while C samples are degraded more than B samples. For DTE25, B, C, and D samples were sampled on August 23, October 30, and January 10, respectively. Different levels of particulate contamination were created in fresh T100 oil by Fe-particles (Yakuri Chemicals, Japan) inclusion (using black-to-gray carbonyl Iron powder Grade-L, mean particle size 7.6 μm). Different levels of oil-ageing were created by thermal degradation (ASTM D2070) in an electric gravity oven.

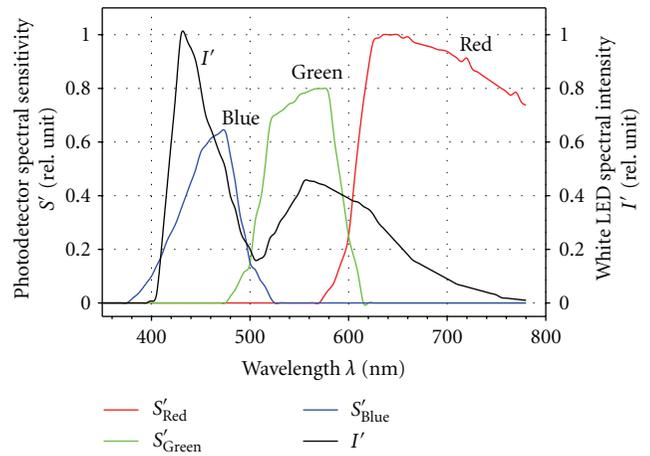


FIGURE 3: Photodetector spectral sensitivity and white LED spectral intensity.

Viscometry (ASTM D445) was performed using Cannon viscometers in a viscometer bath (KV-8, Stanhope-Seta, UK), and TAN tests were performed using kittiwake kits (FG-K25196-KW, FG-K2-006-KW, Kittiwake, UK) configured to ASTM D974/D664. UV-VIS spectrometry (ASTM D275) was performed with photospectrometer (Opron-3000, Republic of Korea).

The detector probe was connected-up to a microcontroller and computer after cleaning; then power was turned on and probe optical windows were immersed in oil sample contained in a vessel. CR, TCI, temperature, and water content (%RS) measurands were recorded. Detector output display range accuracy was ± 0.01 for CR and TCI, $\pm 1\%$ for RS and $\pm 1^\circ\text{C}$ for temperature.

3. Results and Discussion

3.1. *Calibration and Chromatic Processing.* Polychromatic (white LED) source with normalized spectral intensity $I'(\lambda)$ in air (Figure 3) and RGB color sensor with sensitivity $S'(\lambda)$ (Figure 3) was used as photoemitter and photoreceiver, respectively. The color sensor (MCS3AT/BT, MAZet GmbH,

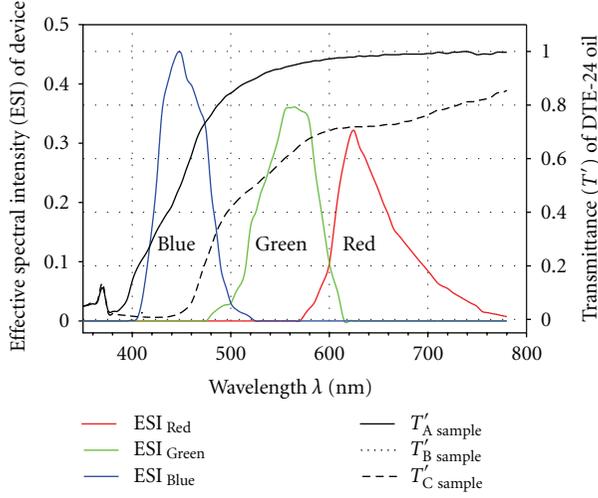


FIGURE 4: Effective spectral intensity and transmittance spectra of DTE24 #1 calibration oil.

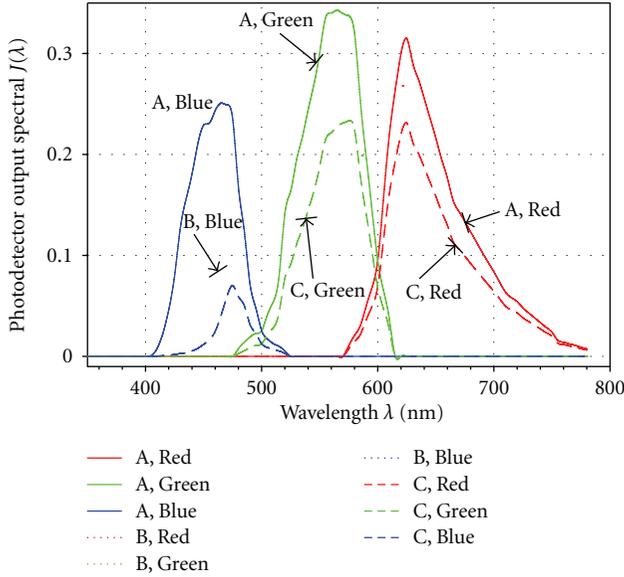


FIGURE 5: Photodetector output spectra in DTE24 #1 calibration oil.

Germany) comprised of 3 Si-PIN photodiodes integrated on a chip. They are carried out as segments of a \varnothing 2 mm ring, with 0.85 mm^2 light sensitive area per segment. Hence, effective spectral intensity $I'_e(\lambda)$ is given by:

$$I'_e(\lambda) = I'(\lambda) \cdot S'(\lambda), \quad (5)$$

where $S'(\lambda) = S(\lambda)/S_{\max}$; $I'(\lambda) = I(\lambda)/I_{\max}$. Photodetector output current (J) becomes

$$J = \int_{\lambda_1}^{\lambda_2} I'_e(\lambda) d\lambda = \int_{\lambda_1}^{\lambda_2} I'(\lambda) \cdot S'(\lambda) d\lambda. \quad (6)$$

The use of normalized parameter values overrides amplifier gain variations. DTE24 #1 oil samples with transmittance T' spectra (Figure 4) measured by multifrequency cross-correlation phase and modulation fluorometer ($K2$, ISS Co.,

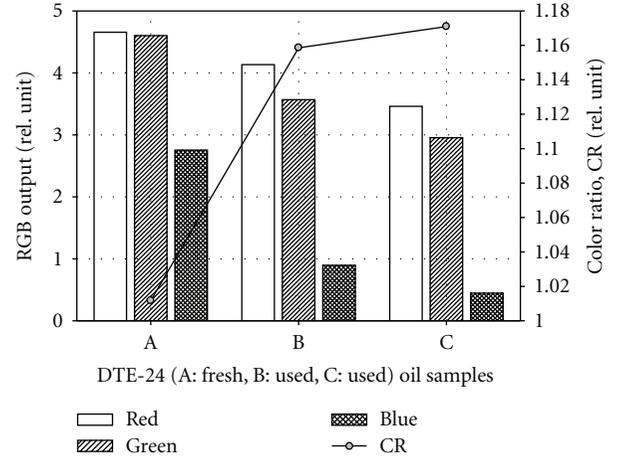


FIGURE 6: Device output in RGB waveband and color ratio (CR) in DTE24 #1 calibration oil.

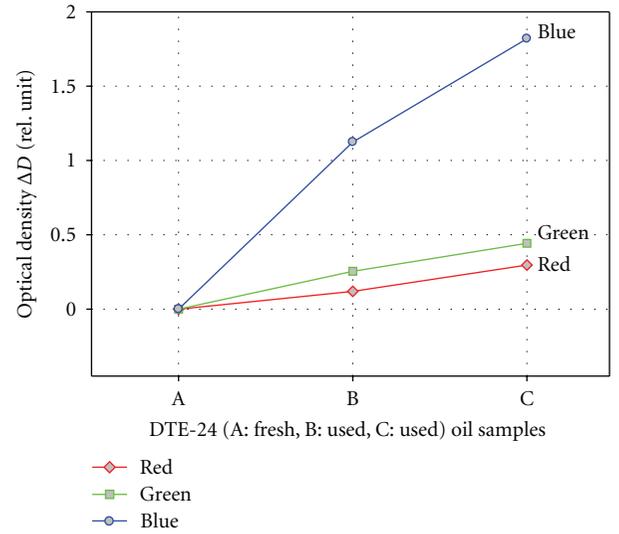


FIGURE 7: Change in optical density of device in DTE24 #1 calibration oil.

USA) was used to calibrate the device. The photodetector output depends on light transmittance through the oil; hence, for calibration of the photodetector output (5) was applied to DTE24 #1 samples to obtain Figure 5. Meanwhile, the RGB output Figure 6 was obtained by integration of the effective spectral intensity I'_e (6) across the entire RGB wavelength ranges. Figure 6 shows that the ratio of output in the Red-to-Green color ratio (CR) increases with oil degradation in use. The optical density referred to as TCI, also increases with the calibration oil degradation (Figure 7).

The outputs were also processed by CIE (Commission Internationale de l'Eclairage) chromaticity coordinates (x, y), Saturation (S) and Hue (H) algorithms of color science [21–23] for comparison. Hence, CR and other chromatic parameters were applied to DTE24 #2 and DTE25 oils sampled a steel factory (POSCO, Korea); the results presented in Table 1. In the chromaticity algorithms equations

TABLE 1: Comparison of CR with conventional chromatic parameters—saturation S , Hue H , CIE—chromaticity (x - y) coordinates, for fresh (A sample) and in-service degraded (B sample and C sample) oils.

Samples	CR	CIE coordinates		Saturation S	Hue H
		x	y		
DTE25 oil					
Air	0.453	0.195	0.430	0.376	165.9
A sample	0.470	0.280	0.596	0.658	79.9
B sample	1.028	0.471	0.458	0.737	58.04
C sample	1.051	0.476	0.454	0.745	56.55
D sample	1.200	0.507	0.422	0.753	48.39
DTE24 #2 oil					
Air	0.453	0.195	0.430	0.376	165.9
A sample	1.012	0.388	0.383	0.257	58.3
B sample	1.159	0.481	0.415	0.644	49.5
C sample	1.171	0.504	0.431	0.771	49.9

(7), Hue H value gives an indication of the dominant optical wavelength, while saturation S value gives an indication of the spread of the spectrum:

$$\begin{aligned}
 x &= \frac{U_R}{U_R + U_G + U_B}, \\
 y &= \frac{U_G}{U_R + U_G + U_B}, \\
 S &= \frac{\max(U_R, U_G, U_B) - \min(U_R, U_G, U_B)}{\max(U_R, U_G, U_B) + \min(U_R, U_G, U_B)}, \\
 H &= 60(k + f),
 \end{aligned} \tag{7}$$

where k is the sector number defined by the order of the magnitudes of U_R, U_G, U_B values; f is the Preucil Hue error or the fraction within the sector of Hue given by the following:

$$f = \frac{\text{mid}(U_R, U_G, U_B) - \min(U_R, U_G, U_B)}{\max(U_R, U_G, U_B) - \min(U_R, U_G, U_B)}. \tag{8}$$

From Table 1, CR parameter like chromaticity CIE- x and CIE- y coordinates obviously increase as oil degradation with service time increases and hence suggestive of a degradation indicator. But, the same cannot be inferred for hue H and saturation S which depend on the $U_R, U_G,$ and U_B , magnitude order. This order changed as sensor location changed from air to different oil samples giving different saturation k values and Preucil Hue error f -values. Figure 8 shows that the device response is stable with respect to time, with CR increasing, and with oil degradation in service.

3.2. Artificially Degraded Oils

3.2.1. Color Ratio (CR) and Total Contamination Index (TCI). In particle-contaminated T100 oil, CR values were practically constant and independent of physical particle concentration as shown in Figure 9. The samples in the figure were purely Fe-particle/oil mixtures without chemical reaction. TCI in

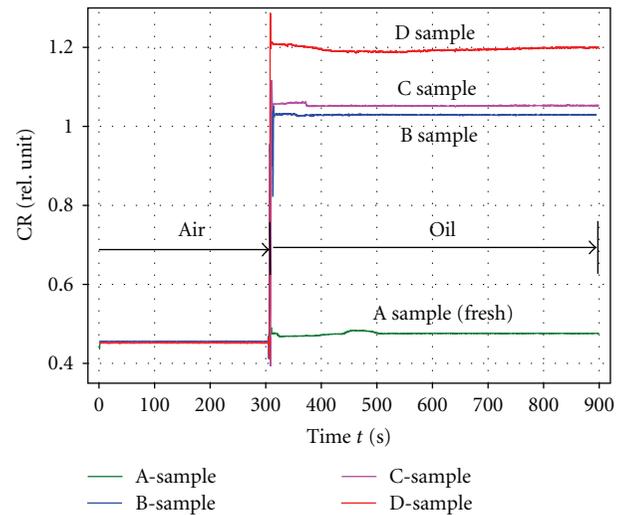


FIGURE 8: CR-time response of detector in DT25 oil for A sample (fresh), B sample (Aug 30), C sample (Oct 23) and D sample (Jan 10).

the 3 wavelengths linearly increased with particle concentration in T100 samples with $R^2 = 0.9956, 0.9925,$ and 0.9961 for the red, green, and blue, respectively. Standard error and sum of square residuals for the regressions were 0.0284, 0.0273, 0.0361 and 0.0073, 0.0067, 0.0117 for the red, green, and blue wavelengths, respectively. Using one-tail t -test, the P value of this regression was P value = $3.294 \times 10^{-12}, 1.917 \times 10^{-12}$ and 3.519×10^{-11} for the red, green and blue waveband respectively; that is a statistical significance of $3.294 \times 10^{-10}\%, 1.917 \times 10^{-10}\%,$ and $3.519 \times 10^{-9}\%$ in the red, green, and blue wavelength correspondingly. Hence, the null hypothesis of TCI independence on physical or particulate contamination in oil is therefore rejected based on less than 0.001% highly statistical significance.

In the thermally degraded T100 samples, TCI increased linearly with increasing ageing time as shown in Figure 10.

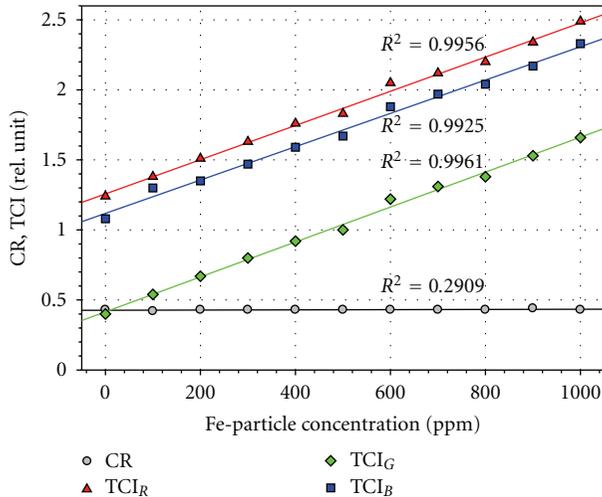


FIGURE 9: CR, TCI variation with Fe-particulate contamination in T100 oil.

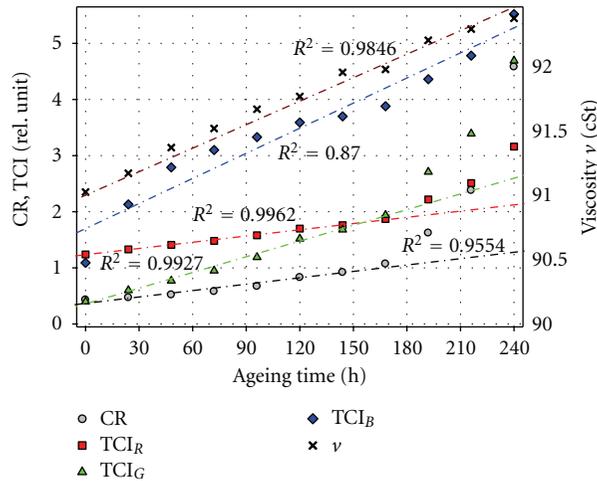


FIGURE 10: CR, TCI and viscosity variation with ageing time of T100 oil.

However, from the 192 h degraded-oil, samples became very dark with CR, TCI_R , TCI_G , TCI_B , and ν_{40} values of 1.62, 2.22, 2.71, 4.36, and 92.24 cSt, respectively; showing a shift from the linearity assumptions of Lambert-Beer's law.

3.2.2. Viscosity and Total Acid Number (TAN). Kinematic viscosity ν (Figure 10) of the thermally degraded oil in this study varied linearly with ageing time, due to the buildup of insolubles and evaporative loss of more volatile components observed by Kaleli and Yavslıli [24]. This variation pattern of viscosity with ageing agrees with the results of Singh et al. [25]. These higher molecular weight insoluble components increase the oil viscosity in the course of oil degradation.

The TAN-values showed little change during thermal degradation, decreasing from 0.20 to 0.15 mgKOH/g after 240 h probably due to the additive depletion either by evaporative loss or decomposition of organic acidic-additive

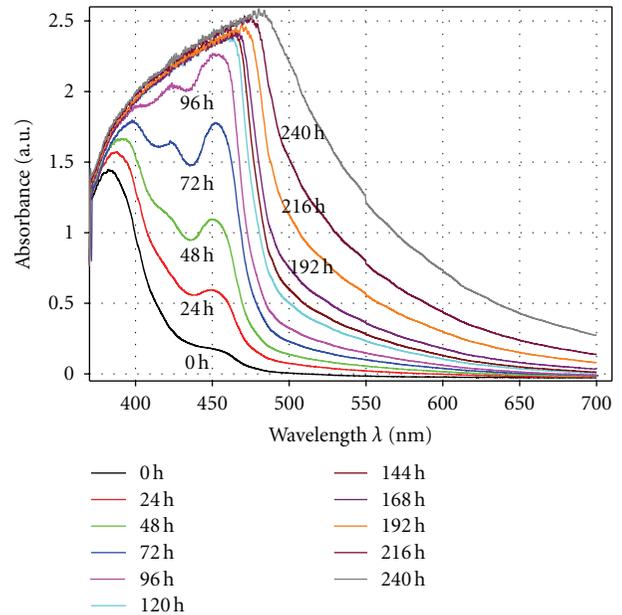


FIGURE 11: UV-VIS optical transmittance spectra of the T100 oil with different ages.

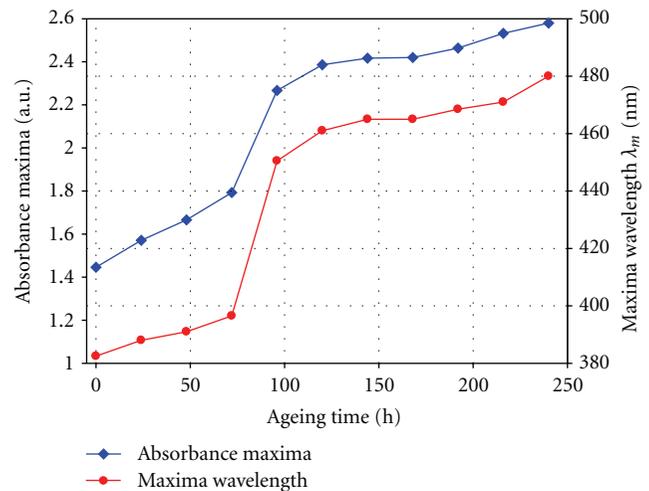


FIGURE 12: UV-VIS absorbance maxima and maxima-wavelength variation with ageing for T100.

in oil, verifiable by molecular spectroscopy [26]. These degraded samples showed CR, TCI_R , TCI_G , TCI_B and ν_{40} values of 4.58, 3.16, 4.69, 5.52, and 92.41 cSt, respectively, for the 240 h sample, and 0.43, 1.23, 0.40, 1.05, and 90.79 cSt, respectively, for 0 h sample. Comparing the change ranges indicates that the CR and TCI were more sensitive to degradation than viscosity and TAN in these samples.

3.2.3. UV-VIS Photospectrometry. The UV-VIS photospectrometry of the artificially degraded samples shown in Figure 11 indicate a steady absorbance increase with increase in ageing time; and a spectral shift from short to longer

wavelength in the course of oil ageing by thermal degradation. Optical maximum-absorbance (absorbance maxima) of the samples and its wavelength of occurrence also increased with ageing time as in Figure 12. Hence, the shift in the absorbance spectra is associated with a shift in maximum-absorbance wavelength from short to longer wavelength. The results of Figures 11 and 12 provide good background support for the measurement principle of the detector.

4. Conclusion

CR and TCI parameters results of the detector were comparable to other color processing parameters of color theory like CIE chromaticity X -, Y -coordinates; and hence, found to be effective criteria for the characterization of the degradation of hydraulic mineral oils. CR depended on oil chemical degradation, while TCI depended on both chemical and physical degradation. TCI was most sensitive in blue waveband and least in the green for the mineral oils. CR and TCI results conform to results of other analyses techniques, such as viscometry, TAN, and UV-VIS photospectrometry.

Nomenclature

A_b :	Absorbance
C :	Concentration of absorption sites
CR:	Color ratio
D :	Optical density
f :	Preucil Hue error
H :	Hue
I'_e :	Effective spectral intensity
I :	Light intensity of emitter
J :	Output light intensity of photodiode
k :	Sector number
k_λ :	Absorptivity coefficient
LED:	Light Emitter Diode
l :	Optical path length
RS:	Relative saturation
S :	Saturation
S' :	Spectral sensitivity
TAN:	Total acid number
TCI:	Total contamination index
U :	Output voltage of the photodiode
X, Y :	Chromaticity CIE Coordinates
ϵ :	Molar absorptivity
ν :	Kinematic viscosity.

Subscripts

i :	Input
o :	Output
R, G, B :	Red, green, blue wavelengths.

Superscripts

f :	Fresh oil
u :	Used oil.

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