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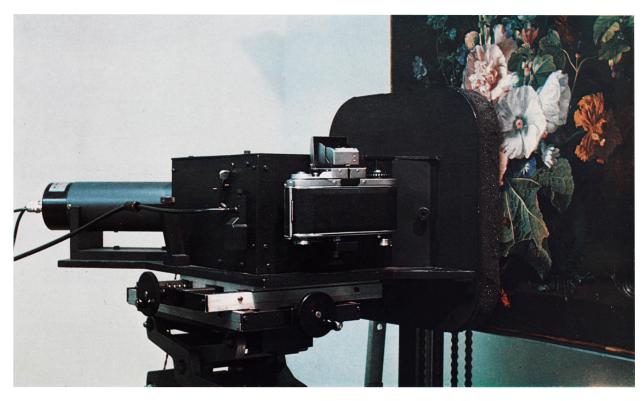


Plate 9 The Wright—Wassall spectrophotometer during measurement of colours of No. 1001, Huijsum, Hollyhocks and other Flowers in a Vase. Conservation reports indicate that a yellow glaze on certain leaves has faded, leaving them a bright blue.



Plate 10 No.6294, Uccello, S. George and the Dragon. The areas of copper resinate green that have been measured are marked. The lower edge of the picture has apparently been protected from light by an earlier frame. See Fig. 5, p.51 for reflectance curves.

Reflectance Spectrophotometry for Measurement of Colour Change

Linda Bullock

The deterioration of artists' pigments has been the subject of a number of studies, one of the earliest being that of Field in 1809 (1,2). Russell and Abney produced a report in 1888 of their very thorough experiments carried out at the request of the Council of Education after a controversy had developed over the introduction of artificial lighting at the then South Kensington Museum, now the Victoria and Albert Museum (3,4). They looked at the effects on watercolour pigments of temperature, humidity, atmospheres of different gases, light intensity and the effects of different parts of the spectrum. Their results are of great relevance and interest as the majority of these pigments were, and still are, used in oil colours, although in this technique the pigment is better protected than in watercolour. Russell and Abney's general conclusions were that light, moisture and oxygen are together essential for fading to occur and that, due to their higher energy, ultraviolet radiation and visible blue light are the most damaging regions of the spectrum.

Little quantitative work has been done on the surface colour of paintings although it is known that changes have occurred, often from comparison of colours adjacent to and below the frame. The important changes induced in oil painting occur with pigments made from organic materials such as the red lake pigments described in the 1977 issue of this Bulletin. One of the most striking colour changes has happened with a pigment known as copper resinate (produced when verdigris or another copper salt is dissolved in a resinous medium), which was widely used during the Italian Renaissance due to the lack of other good greens. It is a bright green glazing colour which has the unfortunate property of changing to an opaque brown under the action of light. Examples of this change are numerous in the Gallery's collection. No. 1034, Botticelli's Mystic Nativity, for example, where the robes of the flying angels were painted with a copper resinate glaze over gold, and No.6307, Giorgione's Sunset Landscape, where the graduation to the distant horizon has been distorted by the abrupt change in pigments.

Other changes also occur with time in the medium and varnish which affect the surface colour of the picture. Natural varnishes gradually change to a dark brownish yellow, obscuring detail and colour, and oils sometimes suffer a loss in hiding power which results in layers below the top surface of paint showing through, such as in No.834, de Hoogh's An Interior Scene and No.3107, 16th century Venetian School, Solomon and the Queen of Sheba.

It is highly desirable to know more about the rates of these changes while paintings hang on exhibition,

rather than under accelerated experimental conditions which can lead to inaccuracies in extrapolation. To this end a programme of work was set up to monitor colour changes occurring on paintings in the Gallery's collection whilst the environmental conditions are also recorded.

The measurement of colour can either be done by colorimetry, which is the specification of colour in terms of the amounts of three primaries, usually red, green and blue light sources, which when mixed will produce that colour, or by the more detailed method of spectrophotometry where the reflectance of a colour throughout the visible spectrum is recorded.

Visual colorimetry was used by Rawlins, the first Scientific Adviser to the Trustees of the National Gallery, for some measurements of colour on paintings in the 1930s. He used a Lovibond Tintometer instrument, first developed for specifying the colour of oils, where a sample colour is matched with a standard lamp whose light is coloured by glass filters (5,6). Amongst other things this apparatus was used to record the colours before and after varnish removal from No.1001, Huijsum's Hollyhocks and No.852, Rubens' Susanna Lunden (?), both of which were exhibited in the 'Cleaned Pictures Exhibition' in 1947 (7). This method, however, is very dependent on the individual observers' colour vision, and the far more detailed and objective method of reflectance spectrophotometry is to be preferred. The first study of artists' pigments using this method was carried out by Barnes (8) on a specially prepared set of panels of uniform, matt pigments, a set of which has passed to the National Gallery Scientific Department from the Fogg Museum in America. The application of these methods to monitor change in museum objects has been discussed (9,10), but until now no long-term study of deterioration has been undertaken.

Because a knowledge of colour changes seemed absolutely basic to a major collection of paintings, the Scientific Adviser, Mr Garry Thomson, decided in the late 1960s to explore various methods by which colour could be recorded with a high enough degree of precision (11). This exploration is still in progress, but the first method is now in use: reflectance spectrophotometry. We were fortunate in being able to enlist the help of Professor W. D. Wright, then of Imperial College, as is described below.

A routine has now been set in motion whereby a group of paintings will have colours at certain points measured every five years. In this way the necessary information on whether and to what degree colours change during normal exhibition conditions will be built up.

Apparatus

There are certain requirements a reflection spectrophotometer must fulfil to be useful:

- 1. It must be capable of great precision and high sensitivity, since the possible colour changes are likely to be small and occur slowly.
- 2. The painting should not come into physical contact with the instrument as its surface may be in a fragile
- 3. The measured area should be of a suitable size for useful information on a uniform colour to be obtainable
- 4. There should be a method of accurately directing the spot of light onto the paint surface under test and of accurately recording the target area to enable measurements to be repeated at intervals in the future.

There was no commercially available instrument suitable for this task, so it was our good fortune that the Paul Instrument Fund provided Professor W. D. Wright with a grant to develop one fulfilling these requirements. The instrument was designed and built with the help of Dr M. P. Wassall and is now on permanent loan to the Gallery.

In principle, white light from a lamp is split up into the colours of the spectrum, blue to green to yellow to red, each is shone in turn onto the area of the painting under investigation and the proportion of the light reflected is recorded (e.g. a strong red colour will not reflect much blue or green light, but will reflect strongly any red light to shine on it). This leads to the required reflectance data through the spectrum.

In more detail, the instrument comprises a monochromator and a measuring head shown diagramatically in Figs. 1 and 2. Light from a tungsten ribbonfilament lamp L enters a large double monochromator consisting of two identical optical systems mounted one above the other, shown schematically in Fig.1. Each system comprises collimating and imaging lenses of two 60° prisms which disperse the white light into a spectrum focussed at a point S. Here a slit of variable width picks out a narrow band of colour which is reflected to the upper system via a frontsilvered mirror. The selected light is returned through the upper system with compensating dispersion, which ensures that the direction of the emerging beam is independent of wavelength and can be brought to a focus at the exit slit E of the monochromator. The wavelength selected can be varied by rotating together the upper and lower prisms P₁,P₂ which swing the spectrum across the slit S. This can be done manually via a micrometer screw or by means of a stepping motor. The waveband selected is controlled by the manually adjusted slit S, and is kept at about 3 nm. The monochromator is just capable of resolving the orange doublet of emission lines from a mercury lamp at 577.0 nm and 579.0 nm, giving it a resolving power of 290.

In practise, reflection readings are required at equal wavelength intervals throughout the visible spectrum, say every 10 nm, from 380 nm at the violet end to 760 nm at the red end. The dispersion of the spectrum

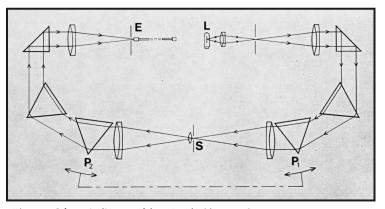


Figure I Schematic diagram of the prism double monochromator which provides light for the measurements of reflectance from the surface of a painting. For clarity a lens is drawn in the diagram after slit S. This is in fact a reflector so that the left-hand prisms and collimating lenses are mounted directly above those on the right. In particular prism P2 is fixed on top of P1 and the pair are rotated as a unit for wavelength selection.

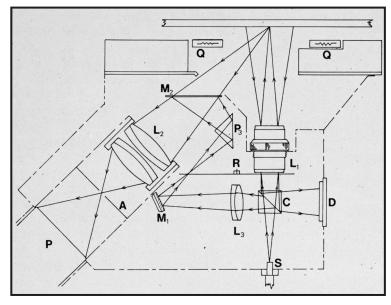


Figure 2 Plan of the measuring head of the instrument which directs the beam of light onto the surface under investigation. The painting is represented at the top of the diagram and the light from the monochromator (Fig. I) enters from the fibre-optics light guide at S.



Figure 3 No.1695, Venetian School, Landscape with Nymphs and Shepherds (see Fig. 4).

by prisms is not linear and the prism must be moved through a greater angle for a 10 nm change in the blue than in the red. To calibrate the wavelength scale a cadmium/mercury/zinc lamp is used to provide sixteen emission lines of known wavelength which can be related to prism position. A computer program interpolates to give the appropriate prism positions at 5 or 10 nm intervals as required.

The beam of light emerging from the monochromator is fed into a flexible fibre-optic light guide which has a slit configuration at one end to match the exit slit, and a circular configuration at the other where it enters the measuring head of the instrument (S in Fig.2). The mounting of this head provides lateral and vertical displacements, together with focussing control to bring the spot of light onto the correct place on the picture, which is held in position on an easel which has motorized horizontal and vertical controls. The light is imaged by a well-corrected lens L₁ onto the area of paint under examination, which measures 4mm diameter. The light that is reflected from the surface is imaged by a lens L₂, whose axis is at 45° to the painting, passes through the aperture A and diverges to fill the area of the cathode of the photomultiplier P, thus ensuring that only light from the sample area can reach the photomultiplier. This configuration of normal incidence and 45° collection is one of the recommended geometries of the C.I.E. (Commission Internationale de l'Eclairage). A beam-splitter C lies between the input S and lens L₁ for two purposes. Firstly, it reflects an image of an area on the painting to the camera at D, which is used both for locating the spot of light onto the correct place using the viewfinder, and for recording photographically its position. Two small lamps Q provide the illumination for this record. The second function of the beam-splitter is to reflect some of the input flux into a secondary optical system L3, M1, M2, P3 taking light to the photomultiplier directly. This system only operates when a lever-operated shutter R blocks light to the picture while allowing light to pass from M₁ to P₃. This gives an internal standard with which to compare the amount of light reflected from the picture. The photomultiplier converts the incident light flux to an amplified electrical signal which is measured on a digital picoammeter (the current usually lies in the range 10⁻⁹ to 10⁻⁶ amps). The output from the picoammeter can be taken to a graph-plotter or to a paper tape punch, the latter being more useful as some computation is required before the final spectral reflectance curve is produced. This computation is performed by the Department's Wang 2200 computer.

Procedure for a typical recording

The painting is examined to select 4mm-diameter sample areas for measurement. Between five and ten areas are usually chosen, either for their likelihood to change, based on our present inadequate knowledge as to their susceptibility, or because they are interesting in some other way. The lightest and supposedly most stable area is also chosen as a check on the yellowing of

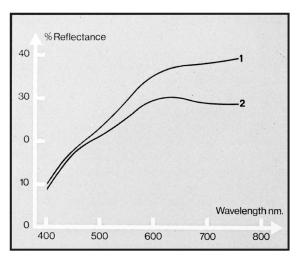


Figure 4 Tracings of reflectance curves from Landscape with Nymphs and Shepherds (Fig.3). Measurements are of the skin colour of the standing nymph above (1) and below (2) the horizon. Below the horizon this is darker because of an increase in paint transparency.

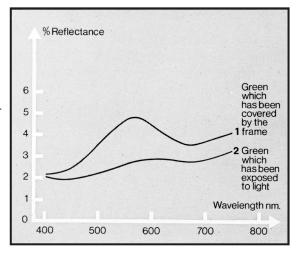


Figure 5 Tracings of reflectance curves from the two sampled areas of copper resinate green on Uccello's S.George and the Dragon (Plate 10, p.48).

the varnish. It should be quite possible to differentiate between a pigment change towards yellow and a varnish change, as the varnish change would affect all the samples in a similar, if not identical way.

The chosen area is located on the instrument and photographed on black-and-white film, with the camera on the measuring head. The visible spectrum is then stepped through at 10 nm intervals (although very occasionally a 5 nm interval is used for extra detail in a particular part of the spectrum) each time the current corresponding to the light flux incident on the picture, the internal standard, and that of the light reflected from the picture is punched onto paper

Once during the day of measurement this process is repeated on a light grey ceramic tile from the set of twelve tiles calibrated by the National Physical Laboratory (12). This is essential and provides an absolute calibration of the instrument, thus taking into account the spectral energy output of the tungsten lamp and the response of the photomultiplier.

The ratio, at each wavelength, of incident to reflected light, weighted by the absolute calibration factors from the ceramic tile, is calculated automatically and a printout is made of the percent reflectance values at 10 nm intervals together with the tristimulus values and chromaticity coordinates for the colours under the standard daylight illuminant D₆₅. See Appendix on p.55.

For each painting examined the following information is recorded:

- 1. Attribution, subject and date of measurements.
- 2. Details on the last treatment to the painting obtained from the conservation dossiers, including varnish type if known.
- 3. Location of the picture in the Gallery, which is updated if it is moved (to link up with environmental records).
- 4. Approximate x,y coordinates of sample points, to nearest 0.25cm, together with any information known about the pigments from microscopical or microchemical analysis.
- 5. A black-and-white photographic negative of each sample point, to be used for future relocation.
- 6. Details pertaining to the spectrophotometer such as lamp type and voltage, photomultiplier voltage, slit widths and wavelength calibration.
- 7. Spectral reflectance values obtained through the visible spectrum, usually every 10 nm together with the calculated tristimulus values and chromaticity coordinates for D₆₅ illuminant.
- 8. A plot of the spectral reflectance curve.

Tracings of the reflectance curves obtained from paintings are drawn in Figs.4 and 5. Future readings will determine whether or not the changes are still going on.

Resolution and repeatability

A thorough study of the precision and repeatability of the instrument was carried out because it is of obvious importance to be able to judge whether or not a measured colour change is real and significant, rather than due to error in measurement. Several precautions have been found to be necessary to increase the accuracy of measurement. The instrument is a singlebeam type and a useful reduction in error is made by taking readings of the sample and internal standard in succession at each wavelength. Due to noise on the signal from the photomultiplier, despite the use of special low-noise screened cable, four readings of the photocurrent are taken each time rather than one, the averaging being done when the computer reads the paper tape. Because the painting is not in close contact with the measuring head but about 1cm away when focussed, the photomultiplier dark current is a significant proportion of the signal; accuracy is improved by reduction of ambient light in the measuring room, particularly at the blue end of the spectrum where the signal is low due to the spectral output of the tungsten lamp. To keep a check on the absolute accuracy of the instrument a set of twelve stable ceramic tiles, which have been calibrated by the National Physical Laboratory, Teddington, Middx., are used as a check over the colour gamut (12). The light grey ceramic standard reflectance is recorded at the beginning of each day's

measurements and any necessary correction factors built into the computer program.

Additional errors are involved, however, when a colour on a painting is remeasured as location of the sample area is involved. For relocation, the black-andwhite negative taken when the area was first measured is replaced in the film plane with the camera shutter and back open. Coarse controls on the easel and head bring the picture to approximately the right position, an image of the painting is then viewed through the negative and the camera system. The fine controls on the head are used to coincide the painting with its negative image, which is visually very precise and obvious, particularly if there is a developed craquelure pattern.

When all the above precautions are taken, analysis of the total error in measurement on a uniform patch of colour, such as the ceramic standards, is about +0.25 per cent in reflectance over most of the visible spectrum, but a slightly higher figure is found at the extremes. The proportional error for dark samples is rather higher. When a painting is repeatedly measured the error in reflectance is nearer \pm 0.5 per cent (see below). The errors this leads to in chromaticity coordinates (x,y) and luminous reflectance (Y) are difficult to assess, as the relationship between the spectrophotometric errors and those in the colorimetric quantities derived from the reflectance data is complex and depends on the shape of the reflectance curve. The probable errors in x,y for measurements on paintings which have been relocated are about 0.002 to 0.005 (depending on their lightness) and the error in Y between about 0.5 per cent and 1 per cent. If a change in colour is measured outside these limits it can be judged a significant one.

Data for evaluating the repeatability of the whole process were obtained from the spectral reflectance curves of a reddish yellow area on an old experimental oil painting (Fig.6). A series of twenty-five complete spectral runs was carried out in this area over a period of six months, each time involving the relocation process with the photographic negative. The errors in reflectance are greatest at the extremes of the spectrum and lowest in the middle. Reflectance values at extremes and middle are listed in Table 1, together with the x,y co-ordinates and luminous reflectance value Y for each run. Also shown are the mean and standard deviation expressed as a percentage of this mean for all the above values. We can be 95 per cent confident that a future run on this sample would lie within the limits $\pm 2 \times$ (standard deviation), if there has been no colour change.

All but one 'rogue' reading (reading no. 4) lie approximately within the MacAdam ellipse for this area of the chromaticity chart. This means that the error in measurement is about the same as would be obtained by the human eye matching sample sets side by side under the best viewing conditions.

It is fair to exclude this rogue reading since in actual practice a variation of this sort would be spotted and the run repeated after relocation.

Table I Repeatability Test									
Run No.	Refl. % at 380 nm	Refl. % at 550 nm	Refl. % at 760 nm	x	γ	Y%			
1	6.0	19.2	53.0	0.438	0.377	24.0			
2	6.5	18.5	49.9	0.435	0.380	21.6			
3	6.4	18.2	50.4	0.434	0.378	21.7			
4	5.7	15.6	43.6	0.424	0.371	19.3			
5	6.8	18.4	52.2	0.436	0.378	22.5			
6	6.2	18.5	52.0	0.433	0.382	22.8			
7	6.4	19.0	52.7	0.436	0.381	22.9			
8	6.6	18.8	52.1	0.433	0.379	23.1			
9	6.4	18.5	51.8	0.436	0.380	22.6			
10	6.3	18.9	52.7	0.435	0.380	23.0			
11	6.2	18.0	50.2	0.434	0.382	22.2			
12	6.8	18.1	51.5	0.438	0.377	22.5			
13	6.4	18.6	52.3	0.434	0.378	22.9			
14	6.0	18.8	52.9	0.435	0.378	22.0			
15	6.0	18.4	51.8	0.435	0.379	22.7			
16	6.5	18.7	51.7	0.436	0.380	22.9			
17	6.2	19.0	51.9	0.434	0.381	23 · 1			
18	6.1	18.8	50.9	0.437	0.377	22.2			
19	6.6	18.3	49.8	0.438	0.378	21.8			
20	5.9	19.0	50.3	0.434	0.378	22.8			
21	6.3	18.4	52.5	0.436	0.380	23 · 1			
22	6.0	18.8	52.0	0.437	0.379	22.4			
23	6.1	19.1	50.9	0.436	0.379	22.5			
24	6.2	18.5	51 · 4	0.435	0.380	22.9			
25	6.5	18.7	51.6	0.436	0.380	22.7			
Mean	6.3	18.5	51.3	0.435	0.379	22.5			
Stand. Dev. %	4.3	3.6	3.5	0.6	0.6	3.7			
Excluding run		40. (F4 6	0.425	0.070	22.6			
Mean	6.3	18.6	51.6	0.435	0.379	22.6			

Figure 6 Repeatability test. Print of the relocation negative used for the twenty-five readings of Table 1. The 4mm-diameter measured area is marked (reproduced

Stand. Dev. % 3.9

1.7



0.3

0.4

2.3



Figure 7 Sunbathers by Neil Thomson.

Fading tests

As an illustration of the possibilities of the instrument, although not as an exhaustive fading test, two experimental pictures were exposed to daylight and ultraviolet light (dosage not measured) the colours being measured both before and after exposure.

Fig.7, Sunbathers, was painted with areas of known stable and fugitive pigments in a gum arabic medium. The stable pigments used were viridian and the quinacridone cinquasia red; those known to be fugitive included red and yellow lake pigments, gamboge and indigo. The pigments used, together with their chromaticities and luminances before and after irradiation are listed in Table 2 and plotted in

There are no significant changes in viridian or the cinquasia red, while the red lakes, gamboge and quercitron all display changes which would be quite noticeable to the eye if the 'before' and 'after' colours could be seen side by side. The spectrophotometer is able to 'remember' exactly and quantify the colour changes. As the plot in Fig.8 is in a uniform chromaticity space, equal distances anywhere on the diagram are approximately equal visual steps. A comparison of the fading of indigo and gamboge gives a clear idea as to how their combined colour would change if it were exposed to light. The fading of gamboge, being a quicker process than that of indigo, would send the green combination towards a blue.

Fig. 9, Art Lovers had a background with yellow and a brownish pink colour merging into one another, the gradual effect produced with an air brush. The pigments used were unknown. The outline detail and some of the foreground colour was done separately on a transparent overlay sheet. After exposure the image of the outline detail was transferred to the background because its colour had faded except on the areas protected by the black lines on the overlay. The outline image was far more noticeable on the pink areas of the background than the yellow, which means that the yellow did not change colour much by comparison, a fact borne out by the reflectance curves in Fig. 10.

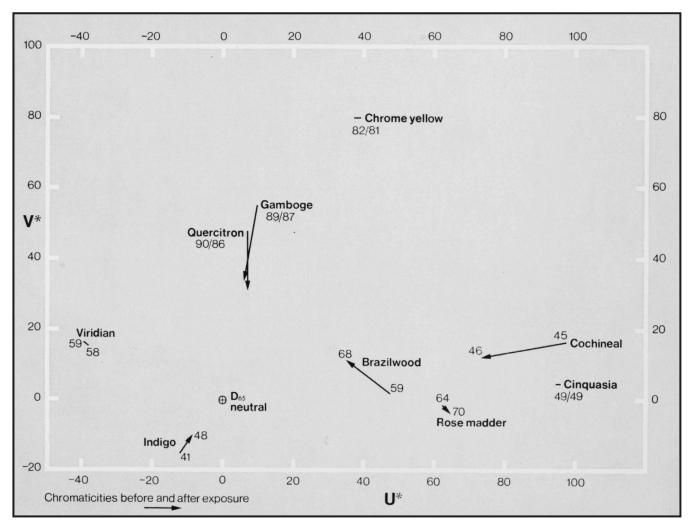


Figure 8 Chromaticity plot of colours in *Sunbathers* (Fig.7) before and after exposure to visible and ultraviolet light. An explanation of the u*v* system used here is found in the Appendix on p.54.

Table 2 Picture A, Sunbathers

Pigment	Origin	Before exposure		After	After exposure		Befor	Before exposure		After exposure			
		x	γ	Y%	х	γ	Y%	u*	v*	L*	u*	v*	L*
Cochineal Lake	NG Lab 18/10/73 Recipe: Marcucci	0.526	0.317	14.7	0.474	0.320	15.3	98.89	16.29	45.2	74.33	12.27	46.0
Brazilwood Lake	NG Lab 23/7/74 Recipe: Tingry	0.387	0.311	26.6	0.374	0.334	37.5	47.35	1.13	58.6	36.31	10.46	67.6
Chrome Yellow	Winsor and Newton	0.449	0.464	59.5	0.445	0.465	58.8	38.66	80.77	81.6	35 · 64	80 · 13	81.2
Viridian	Winsor and Newton	0.258	0.389	26.6	0.261	0.387	25.5	-40.70	16.20	58.6	$-39 \cdot 26$	15.83	57.5
Cinquasia Red	RT-759-D(PS-63011) du Pont	0.482	0.293	17.8	0.485	0.293	17.6	95.83	4.27	49.3	95.97	4.57	49.0
Rose Madder	Winsor and Newton	0.395	0.298	32.9	0.388	0.296	39.2	62.81	-3.97	64 · 1	63.61	-6.34	68.9
Natural Indigo	Winsor and Newton	0.262	0.290	11.9	0.275	0.303	17.3	-11.61	-16.07	41 · 1	-10.72	<i>−</i> 12·79	48.7
Gamboge	Sample from 1851 Exhibition	0.372	0.414	74 · 1	0.349	0.379	69.6	9.57	54.92	88.98	6.87	33.46	86.8
Quercitron Lake	Winsor and Newton (1937)	0.363	0.401	76.4	0.349	0.376	68.3	8 · 44	48.01	90.05	8.03	31.72	86.2

Figure 9 Art Lovers by Neil Thomson.

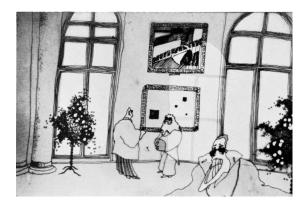
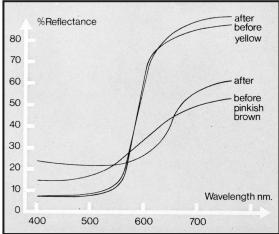


Figure 10 Reflectance curves of yellow and pinkish brown background colours on Art Lovers (Fig.9) before and after exposure.



Summary

The reflectance spectrophotometer at the National Gallery is now being used for routine measurements of the surface colours on paintings in the collection to monitor any colour changes which may be slowly taking place. It is intended to repeat the readings at five-year intervals and to relate the results to the recorded information on the environmental conditions in which the pictures have been hanging.

Appendix: computation of C.I.E. tristimulus values and chromaticity co-ordinates

The tristimulus values X,Y,Z are given by the equations:

$$X = k \sum_{\lambda} r_{\lambda} \overline{x}_{\lambda} H_{\lambda} \Delta \lambda$$
 $Y = k \sum_{\lambda} r_{\lambda} \overline{y}_{\lambda} H_{\lambda} \Delta \lambda$ $Z = k \sum_{\lambda} r_{\lambda} \overline{z}_{\lambda} H_{\lambda} \Delta \lambda$

where,

 r_{λ} is the spectral reflectance of the object.

 $H_{\lambda}\Delta\lambda$ is the spectral distribution of the flux irradiating the surface.

 $k={
m constant}, {
m chosen}$ to give $Y={
m 100}$ for perfect white diffuser.

 $\overline{x}, \overline{y}, \overline{z}$ are the colour-matching functions of the eye which can either be the C.I.E. 1931 standard observer (for fields up to 4° angular subtense) or the 1964 large-field standard observer. See (13,14).

The chromaticity co-ordinates are x, y, z, where:

$$x = X/(X+Y+Z) \quad y = Y/(X+Y+Z)$$
$$z = Z/(X+Y+Z)$$

Only the x,y co-ordinates are required to specify the surface colour and its Y value is the luminous reflectance expressed as a percentage.

In 1976 the C.I.E. adopted a new co-ordinate system which provides a colour diagram much more closely related to the eye's visual discrimination across the colour diagram. In other words, equal distances between colours plotted on the colour diagram represent equal visual steps, an important consideration when trying to judge the significance of changes in colour.

The new co-ordinates are:

$$u^* = 13 L^*(u'-u_n')$$
 $v^* = 13 L^*(v'-v_n')$

and L^* for luminance

where, $L^{\star} = 116(Y/Y_n)^{1/3} - 16$

$$u' = 4X/(X+15Y+3Z)$$
 $v' = 9Y/(X+15Y+3Z)$

 u_n' , v_n' are u', v' for illuminant colour.

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