

The Importance of Chromaticity in the Evaluation of Whiteness¹

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FOR over two decades it has been recognized that the grading of nearly white textile fibers requires the quantitative measurement of their color. As early as 1931 Nickerson (12) described procedures for the grading of raw cotton, by means of disk colorimetry, which provide an adequate description of the color of the samples. MacAdam (11) subsequently examined the color range typical of laundered goods and the relative influence of yellowness and of grayness in the subjective evaluation of whiteness. Judd (7) extended these studies in considering the whiteness of paper samples, by including not only neutral and yellow tinges but also blue ones.

In the finishing or laundering of white cotton goods it is one of the main objectives to render them as white as possible; the ultimate basis of judgment is the visual appearance of the treated or laundered cloth. The evaluation of scouring agents and detergents by end-use tests is however a lengthy and expensive procedure. Therefore laboratory tests have been sought which will provide an adequate rating of the relative merits of detergents. No test has been devised to date which corresponds sufficiently closely to the results obtained by observers judging actual laundered goods. The basis of such visual observations is not the cleanness of the clothes but their whiteness.

Laboratory tests of detergent efficiency, on the other hand, have always been based on measurements of reflectance, or lightness of the washed swatches, a quantity which does not provide an adequate measure of whiteness. Through the consideration of the factors influencing judgments of whiteness it is possible to define the requirements for an improved objective rating of laundered cloth samples. At the same time this improvement in accuracy of the laboratory methods clarifies the need for soiling procedures which are realistic with regard to the ways in which they may impair whiteness.

The Objective Description of Whiteness

The Color of Nearly-White Samples. A nearly-white sample may deviate from perfect whiteness in two ways: it may be grayish, or it may be chromatic. If it is gray, it reflects the same fraction of the incident light at all wavelengths, and its color properties are fully described by reflectance alone. If it is chromatic, then three quantities are required for a description of the sample's color. The chromaticity must be described by stating the hue and the saturation of the color; in addition, it is necessary to describe to what level of lightness (on a gray scale) that color corresponds.

In the subsequent discussion these properties of color will be described in accordance with the standard CIE* system of color specification, which was

formerly known as the ICI system. It will also be useful, on some occasions, to refer to the Munsell system which simplifies the interpretation of color specifications in terms of visual effects. For the detailed description of these systems, the reader is referred to a standard treatise (8).

In the CIE system chromaticity is described by the rectangular coordinates x and y , and visual reflectance by the Y -coordinate. (This visual reflectance, or an approximation to it, is the quantity determined in all published work on detergency testing.) Chromaticity may alternatively be described by Munsell hue and chroma, or, in the CIE system, by dominant wavelength λ_D and excitation purity p_e . Since yellowness is of particular importance in detergency problems and yellow samples necessarily fall within a narrow range of λ_D around 575 $m\mu$, it suffices to specify solely the excitation purity. Excitation purity varies from zero for a gray sample to unity for the most saturated color.

The determinations of the spectrophotometric reflectance and of the CIE psychophysical color specifications given in this paper were made with the G. E. Recording Spectrophotometer, equipped with the GAF-Librascope Tristimulus Integrator (3). The integrations, necessary for obtaining the tristimulus values (X , Y , Z), are

$$\begin{aligned} X &= \int_{\lambda_1}^{\lambda_2} E\bar{x}Rd\lambda, \\ Y &= \int_{\lambda_1}^{\lambda_2} E\bar{y}Rd\lambda, \\ Z &= \int_{\lambda_1}^{\lambda_2} E\bar{z}Rd\lambda, \end{aligned} \quad 1$$

where E is the spectral distribution of energy in the illuminant under which the color is viewed, \bar{x} , \bar{y} , and \bar{z} are the tristimulus values for the spectrum colors, R is the spectral reflectance of the sample, λ is the wavelength, and λ_1 and λ_2 are the limits of the visible spectrum. CIE Illuminant C, which corresponds closely to average daylight, was used in all the automatic computations. At this point it should also be mentioned that the trichromatic coordinates (x , y) are obtained by expressing X and Y as decimal fractions, viz.,

$$x = \frac{X}{X + Y + Z} \quad \text{and} \quad y = \frac{Y}{X + Y + Z} \quad 2$$

The excitation purity (p_e) can be readily obtained by plotting the x and y values on the chromaticity diagrams published by Hardy (6).

Practical Examples of Whites. The use of blueing dyes to improve whiteness of off-white fabrics is a commonly accepted practice. It is based on the cir-

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* CIE is the abbreviation for Commission Internationale de l'Éclairage.

cumstance that off-white samples are most commonly yellow and that a blue dye, by counteracting the initial yellowness, improves the subjective impression of whiteness even though the reflectance of the sample is simultaneously reduced. These circumstances can be usefully described with the aid of Figure 1, which

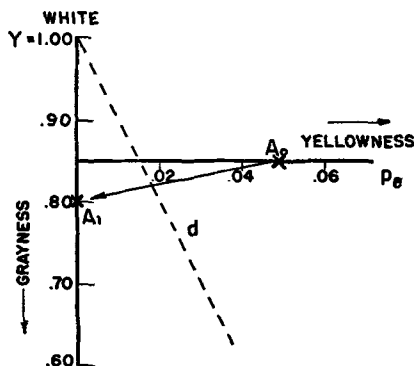


FIG. 1. Diagram of gray and yellow near-whites.

is a schematic representation of gray and yellow near-whites. The vertical axis represents a scale of reflectance, or Y-value, with the perfect white at the top and grays falling in the axis. The horizontal coordinate is the excitation purity (p_e) at $575 \text{ m}\mu$; the point marked A_0 represents a typical slightly yellow material, with coordinates $Y = 0.85$, $\lambda_D = 575 \text{ m}\mu$, and $p_e = 0.05$. If sufficient blueing dye is added to this sample to neutralize fully the initial yellowness, a sample can be obtained with characteristics shown at point A_1 on Figure 1. Here the excitation purity is zero; the reflectance, or Y-value, has been reduced from 0.85 to 0.80. Observers of these two samples will without exception describe the second sample as the whiter, in spite of its lower reflectance, or lightness. If an observer is asked which of these two samples is "brighter," he can likewise normally be relied on erroneously to select the second sample since visual observation does not necessarily permit one to distinguish between brightness and whiteness. For further discussion of this point see Evans (5).

The foregoing circumstances may seem, at first sight, to lead to irreconcilable conflict between whiteness judgments and the use of objective color specifications. No such conflict exists; the procedures for resolving the apparent anomalies are described in the following section.

Factors Influencing Whiteness of Laundered Goods. The factors influencing the subjective impression of whiteness were first systematically investigated by Judd (7). His studies were based on the judgments of colorists trained in the evaluation of white papers. While this analysis is equally applicable to the evaluation of whiteness of laundered goods, it is not to be assumed that the criteria employed by paper colorists are identical to those used in judging white textiles. Judd's work has recently been supplemented in Holland by the studies of Selling and Friele (17, 18); working with textile samples, they have independently reached similar conclusions. In the following discussion one of the formulas for evaluation of whiteness most recently given by Judd (8a) will be employed.

This formula provides a basis for evaluating the relative degradations of whiteness caused by chromaticity and by grayness. Since the primary cause of degradation of whiteness in detergency is yellowness, a simplification of Judd's formula is useful, obtained by reference to the graphs in Hardy's Handbook (6). The modified formula, applicable only to yellow samples of dominant wavelength about 575 to $580 \text{ m}\mu$, is

$$W \cong 1 - \sqrt{25 (p_e)^2 + \left[\frac{1 - Y}{2} \right]^2}, \quad 3$$

where W is the whiteness of the neutral or slightly yellow sample, p_e is its excitation purity, and Y is its visual reflectance, or lightness.

The scale of whiteness defined by equation 3 is such that the optically perfect³ sample, exhibiting 100% reflectance throughout the visible spectrum, is of unit whiteness. A sample of excitation purity 0.2, or Munsell chroma about 2, is of zero or negative whiteness, no matter how high its reflectance may be. Typical nearly-white textile materials are found to have whiteness ratings between 0.4 and 0.9 on this scale. In order to obtain a better comprehension of the significance of the scale, it is instructive to determine the whiteness ratings of the samples specified in Figure 1. The original sample, A_0 , has a whiteness rating $W = 0.74$; the sample A_1 , treated with blueing dye is characterized by $W = 0.90$. This conforms well with visual observations.

To those familiar with the Munsell system of color specification, it is instructive to observe that equation 3 leads to the conclusion that an increase of yellowness by 0.1 Munsell chroma unit is about as detrimental to the visual impression of whiteness as a decrease of 0.5 Munsell value unit. (In the region of near whites 0.5 Munsell value unit is equivalent to 0.1 change in Y value, or 10% change in reflectance.)

It is possible, with the aid of equation 3, to determine under what circumstances the degradation of whiteness is produced primarily by grayness and under what circumstances yellowness is primarily responsible for the visual departure from whiteness. This is indicated in Figure 1, in which the dotted line d represents the specifications of samples for which the degradation of whiteness due to grayness is equal to that due to yellowness. Samples lying between the line d and the Y-axis are predominantly influenced by grayness; those lying to the right of the dotted line are mainly influenced by yellowness. Only if samples lie very much closer to the Y-axis than to the dotted line, can the yellowness be neglected in the evaluation of whiteness.

The only published data on the color of laundered materials is contained in MacAdam's (11) early paper. While his results must be supplemented by more detailed observations of appropriately selected samples, they provide a preliminary basis for examining the relative importance of grayness and of yellowness in laundered goods. Of 23 samples for which he presents data, only four were of such colors that the degradation of whiteness due to grayness exceeded that due to yellowness.

³ Judd has pointed out that some observers might prefer a chromatic white (e.g., slightly bluish sample) to the "optically perfect" white thus defined; he also showed that this case can be precisely treated by a shift of coordinates in the algebraic formula for whiteness.

Laboratory Evaluation of Detergents

The Color of Materials in Detergency Testing. In the various conventional soiling procedures for textiles which precede the laboratory washing operations, carbon black has been the sole or predominant component in the soiling mixtures. The limitations of these artificial soils were enumerated in detail by Lambert and Sanders (9); furthermore, since carbon black is a neutral soiling agent, it imparts no chromaticity to the treated fabric. This is demonstrated in Table I and Figure 2 by the color specifications ob-

TABLE I
Color Specifications of Bleached Cotton Soiled with Varying Amounts of Molacco Carbon Black

Sample	x	y	Y
Original cloth.....	.3114	.3191	.863
Low soil content.....	.3111	.3190	.798
Intermediate soil content.....	.3111	.3187	.746
High soil content.....	.3104	.3177	.574

tained for a series of cotton swatches soiled with Molacco carbon black. De-sized English broadcloth was used in this and the following experiments after it had been washed in a Launder-Ometer with soap, sodium carbonate, and sodium hypochlorite according to AATCC Standard Method 36-45 (1). It was found that this treatment removed essentially all finishing and blueing agents normally present in cloth of this type. The carbon black and also the other soils (see below) were applied by means of a modified Schiefer Abrasion Meter, following the techniques described by Sanders and Lambert (16).

It is evident from the diagram shown in Figure 2 that the increasing grayness (or decreasing lightness) with added amounts of this carbon black is not accompanied by any build-up of color such as is typical of actual laundered goods. In fact, such color as was initially present is masked by the presence of this neutral soiling agent; this is shown by the shift of the points characterizing the color of the soiled swatches toward the neutral point ($x=0.3101$; $y=0.3163$).

In contrast, it is of interest to observe the color specifications for cotton samples which were soiled with various natural soils whose physical-chemical characteristics were described previously (16). The results are given in Table II and also Figure 2. A sample containing enough natural soil to reduce the reflectance from an initial 83% to 73% is measurably yellow; the yellowness is described by an excitation purity of 0.03. According to formula 3, the degradation of whiteness arising from chromaticity is under these circumstances about twice as great as that arising from grayness.

The spectrophotometric reflectance curves for two typical swatches, one soiled with carbon black, the other with natural soil, are shown in Figure 3. The curves shown are for different quantities of soil which

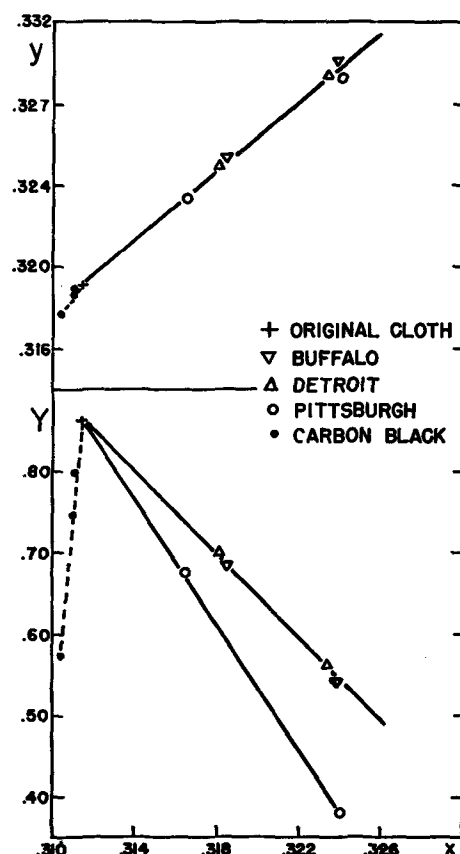


FIG. 2. Color specifications of cotton soiled with natural soils and carbon black.

were chosen so that the visual reflectances would be about equal. These curves are useful for a consideration of their radically different shapes. The very flat curve for the carbon black soil eliminates the necessity, in the instrumental evaluation of cloth soiled with it, of any critical selection of the filter-photocell system employed in the measuring photometer. On the other hand, if a more realistic soil is applied, with a spectral selectivity such as is shown by the curve obtained for the other swatch, it becomes necessary to consider the color as well as the visual reflectance. For an adequate instrumental analysis by means of filter photometers, it is then essential to select filter-photocell systems which conform accurately to an appropriate system of color specifications.

Early laboratory work on detergency evaluation, taking into account also chromatic effects that could influence performance ratings, was described by two investigating teams (2, 15) which visited two factories of the I. G. Farbenindustrie in 1945. In the routine cotton- and wool-washing tests natural or synthetic road dust was applied to the test fabrics from a carbon tetrachloride suspension. In this case not only the natural but also the synthetic soiling mixture im-

TABLE II
Color Specifications of Bleached Cotton Soiled with Varying Amounts of Natural Soils

Sample	Buffalo			Detroit			Pittsburgh		
	x	y	Y	x	y	Y	x	y	Y
Original cloth.....	.3110	.3187	.863	.3110	.3187	.863	.3110	.3187	.863
Low soil content.....	.3184	.3253	.683	.3181	.3250	.697	.3165	.3234	.675
Higher soil content.....	.3238	.3301	.542	.3234	.3295	.563	.3241	.3293	.580

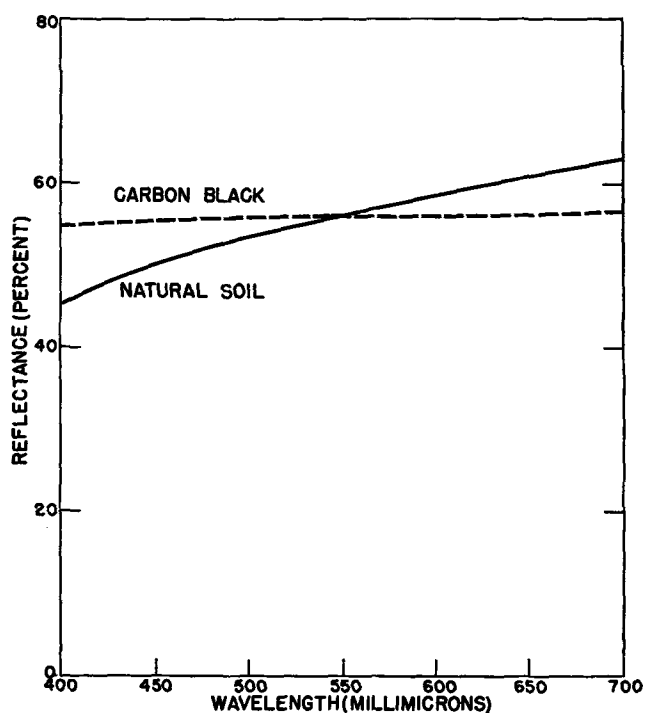


FIG. 3. Spectrophotometric reflectance curves of soiled cotton.

parts chromaticity since the latter contains 1% yellow iron oxide and 2% black iron oxide in addition to lamp black. The visual judgments which were apparently found necessary to supplement the data obtained with a photometer might serve as indirect evidence that chromatic effects were observed and found of importance in this work.

An interesting trend in the application of natural soils in the field of textile testing is illustrated by the recent study carried out by a committee of the AA-TCC (10), in which the soiling of fabrics was evaluated by realistic laboratory methods. However the photometric methods used for the evaluation of the soiled samples were not adequate for detecting the chromatic effects which must have been present; the reflectometer measurements and the black-and-white reproductions shown in the publication describe the soilings obtained merely on a relative scale without due consideration of the visual effects. Many years ago Ringeissen (13) made some experiments with natural soils in detergency testing by using processed sludges from dry-cleaning plants. In a very recent paper (14) the same author emphasizes the importance of colored materials in the soiling and subsequent laundering of garments; nevertheless in the laboratory methods described in the same publication the predominant role of yellowing materials is not at all considered, since oil-soluble green and blue dyes are recommended for determining the degreasing efficiency of soaps and detergents.

In the attempt to devise a more realistic cotton detergency test a multi-component synthetic soil was formulated by Sanders and Lambert (16) which is similar in its chemical and physical characteristics to natural soils. Various batches of this soil prepared by different operators over the period of several years were examined with regard to uniformity of the chromatic effects which they impart to samples soiled by

them. The coloring tendency of five randomly selected batches varied; some soils were exactly equivalent to the natural Buffalo and Detroit soils while others imparted appreciably more color to the cloth. The main coloring constituents in these synthetic soil mixtures are humus, which probably will vary in its color from lot to lot and the amber-colored iron oxide, which is presumably controlled within much narrower tolerances.

It was found that the synthetic soil samples of highest coloring efficiency impart definitely more yellowness to the cloth than did the natural soils described above. In this case a sample containing enough soil to reduce the initial reflectance by 10% has an excitation purity of 0.055. The degradation of whiteness due to chromaticity is under these circumstances about six times as great as that due to grayness. The soil sample used in the experiments described in the following section was one of such high coloring efficiency.

With the three types of soiling materials, carbon black, natural, and synthetic soil, which were under consideration, a wide range in the relative importance of grayness and of color in impairing whiteness has been found. For carbon black all the degradation of whiteness is due to grayness; for other soils, more typical of actual ones, only a small part, in some instances a negligible part, is due to grayness. Under these circumstances the measurement of only one reflectance in the laboratory evaluation of detergents provides a measurement of an aspect which is relatively unimportant in the subjective impression of whiteness. The laboratory test of reflectance, even though it is not measuring the most important quantity, would nonetheless provide an accurate rating of the relative merits of detergents, providing that all have the same influence on the color of the laundered goods.

There are however a number of ways in which detergents may exert a selective influence on the removal or deposition of colored materials. They may differ in their ability to remove the yellowing ingredients of common soils; they may differ in their tendency to permit the gradual build-up of colored impurities present in the wash or rinse waters, and they may differ with respect to the intrinsic yellowing tendencies of any detergent remaining in the cloth. If any such differences exist, it becomes necessary to consider chromatic characteristics in addition to reflectance for a reliable description of the visual whiteness. The subjective evaluation of whiteness is so sensitive to minute degrees of yellowness that even slight differences may well be of controlling importance.

Laboratory Wash Tests with Chromatic Soil. In order to illustrate the existence of selective detergent action, some preliminary observations are presented here which were made in the course of a related study. The colors of cotton broadcloth swatches were measured after they had been laundered in a realistic laboratory wash test (16), using three different detergents and hot water (60°C.) of 100 p.p.m. hardness as a reference. Although the designations of the washing compounds (i.e., C—soap and sequestering agent, D—built anionic, and E—nonionic surfactant) are the same as those used previously (9), the products were not identical but merely similar. Before washing, the swatches were soiled with a special synthetic

soil which contained C^{14} -labelled carbon black instead of the Molacco black, which has been commonly used in the past. The details of the detergency studies with radioactive soils will be described elsewhere.

The color specifications of the laundered swatches, after one cycle of soiling and washing, are shown in Figure 4. The points characterizing the chromaticity

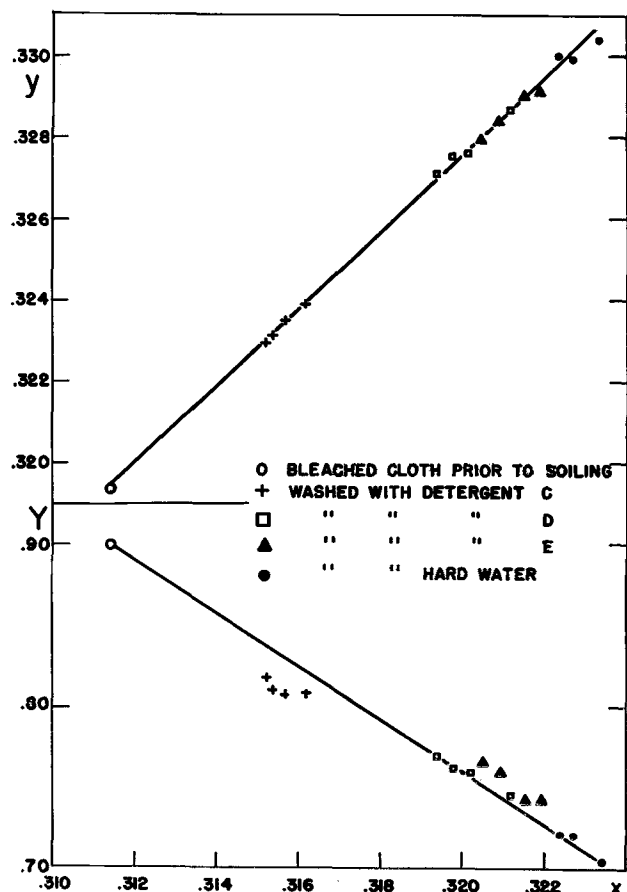


FIG. 4. Color specifications of washed cotton soiled with synthetic G.A.&F. soil.

of the various samples fall closely on the line of dominant wavelength, $\lambda_D = 576 \text{ m}\mu$. There exist however significant deviations of two groups of plotted points from a straight line in the diagram of Y-value vs. chromaticity coordinate x. On the basis of these re-

sults it can be concluded that in comparison to water alone (or to Detergent D) more yellowing of the fabric was produced with Detergent E and less with Detergent C than would correspond to uniform soil removal. Supporting evidence for such effects has been obtained by the use of synthetic soils containing various radioisotopes; it was found that detergents vary considerably in their detergency efficiency for carbonaceous (black) and miscellaneous (yellow) soil constituents. It follows from the foregoing that a comparison of Detergents D and E, based on the measurement of reflectance alone, would lead to a spuriously high rating of Detergent E.

That increased resolution can be attained by chromaticity measurements is illustrated in Table III, where those data of Figure 4 are tabulated which refer to Detergents D and E. The individual values obtained in four replicate tests and the averages of the measured quantities, Y and p_e , are listed. The standard deviations, s_w , and the 95% confidence intervals were directly determined from the respective ranges by use of the simplified statistics for small numbers of observations proposed by Dean and Dixon (4). The range, w, is equal to the difference between the maximum and minimum value, identified by asterisks for each set of data. Although the difference between the average Y-values for both Detergents D and E is equal to the difference between the average p_e -values (i.e., 0.006), the superiority of D over E is indicated only by the chromaticity determinations. A statistical analysis using the *t* test shows that while Detergent D is superior to Detergent E at the 10% level of significance on the basis of the chromaticity data, no significant difference is obtained when the reflectance data are tested.

Conclusions

The factor of paramount importance in the visual evaluation of whiteness of laundered goods is yellowness. This yellowness has in practice been neutralized in various ways, first by the use of bleaches and/or blueing dyes and more recently by means of fluorescent dyes. Any such additive provides at best however an imperfect solution to the problem of obtaining maximum whiteness. Bleaches may cause fiber damage, and blueing dyes are undesirable because their absorption prevents attainment of maximum whiteness; fluorescent dyes are not permanent because of their limited light-fastness and, more fundamentally, because a sample containing sufficient fluorescent dye to be neutral under a given illumination

TABLE III

Illustration of Increased Resolution Afforded by Chromaticity Measurements					
Detergent	Test No.	Reflectance Data		Chromaticity Data	
		Y	Range (w), Stand. Deviation (s_w)	p_e	Range (w), Stand. Deviation (s_w)
D	1	.761	w = .024 s_w = .012	.055	w = .009 s_w = .004
	2	.745*		.062*	
	3	.758		.056	
	4	.769*		.053*	
	Average	.758 ± .017		.056 ± .0065	
E	1	.759	w = .024 s_w = .012	.061	w = .007 s_w = .003
	2	.742*		.064	
	3	.743		.065*	
	4	.766*		.058*	
	Average	.752 ± .017		.062 ± .0050	

will necessarily appear colored when viewed under another light source containing a different ratio of ultraviolet to visible light. It is therefore essential, in the search for the best detergents, to evaluate them first in the absence of any additives and to employ bleaches, blueing, or fluorescent dyes only to minimize their limitations.

The formulation of suitable soiling procedures and the application of reliable methods for measuring the washed cloth samples represent two important steps in the development of satisfactory laboratory tests for evaluating detergents. These steps are necessarily related to one another in a fundamental way. The procedures used in the past for detergency evaluation, involving the measurement of reflectance alone, have been appropriate with the soiling methods relying on carbon black as primary soiling agent but are not useful if realistic soiling techniques are employed and the degree of whiteness is to be determined.

While the colorimetric principles for an adequate objective rating of whiteness have been developed, the actual working methods should be based on a modern re-evaluation of the relative importance of the various colorimetric and psychological factors related to visual estimates of the whiteness of textiles. Such a re-evaluation, coupled with more appropriate soiling and measuring techniques, will lead to improved procedures for the laboratory evaluation of detergents.

Acknowledgment

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The Intra-Molecular Hypothesis of Stand Oil Formation

Letter to Editor:

POWERS (5) has criticized the conclusions drawn by Joubert and Sutton from their study (4) of the composition of a pilchard stand oil. It is not the writer's intention to become involved in a polemical discussion, but it is, on the contrary, worthwhile to consider the points raised to see how far they are met by the available data and to discuss what further work is needed.

The central issue involved is whether triglyceride oils are, on heating, first predominantly converted to intrapolymers¹ (i.e., molecules of the same weight but having a direct linkage between two fatty acid chains) or whether the first predominant reaction is linkage between two chains of different triglyceride molecules. Powers had deduced the former hypothesis whereas Joubert and Sutton's interpretation of their results supported the latter. The method employed was to separate from a stand oil that fraction having no increased molecular weight ("unpolymerized triglycerides") as compared with the original unbodied oil; the fraction was then saponified and separated by molecular distillation into monomeric and higher esters. The total polymers ("polymerized triglycerides") were also examined.

Powers states that molecular distillation (one-stage) is not a highly efficient separation method. We were however faced with the easy separation problem of separating only partially the monomeric esters from dimeric esters of about twice their molecular weight.

¹It is assumed that references to "interpolymers" in Powers' letter are typographical errors and that "intrapolymers" should be substituted.

In fact, we now know (2) that the same still separates methyl linoleate dehydro-dimer from methyl linoleate in a single passage so that considerably less than 2.5% of the former co-distills with the latter. In the pilchard stand oil work we did not require a perfect or near-perfect separation since the monomer:dimer ratios were simply calculated from the molecular weights of those fractions containing species higher than monomers. As is usual and necessary in this calculation, it was assumed that the other species present were dimers or trimers. The saponification equivalents were necessary because pilchard oil contains acids extending over a range of chain lengths. They were not by themselves used as an "index of the degree of polymerization." All assumptions made in the paper were clearly stated.

POWERS' statement that pilchard stand oil may be expected to be essentially different from linseed stand oil in respect of intrapolymer formation is hardly borne out by the data available (6), which have recently been reinforced by the work of Boelhouwer, Jol, and Waterman (1). Further work suggested is the examination of a range of oils bodied to different viscosities, but the labor involved would be very considerable.

The esters of the polymerized triglyceride fraction should contain, as Powers states, at least 33% of dimer acids, and this was stated in the paper. In fact, the figure observed was 33%, after making allowances for hold-up losses. As was pointed out, such losses in