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Thermochromic materials in the paints and varnishes sector. Part I: perception and measurement of color

Materiały termochromowe w sektorze farb i lakierów. Część I: postrzeganie i pomiar barwy

Considerations about thermochromism should begin with exploring the issue of color. Only the assimilation of the basic laws and phenomena that rule the world of colors, and the resulting classification systems, will allow for a proper understanding of this matter. This article presents the most important information regarding color perception, especially from a mathematical perspective, in a concise and accessible way. Color spaces known and used in the paint and varnish sector are described, including: CIELab, and commercially important color classification systems, such as RAL or Natural Color System. The methods of color measurement along with dedicated equipment and standards that are used in the everyday work of a coatings technology specialist are presented. A brief review of the state of knowledge in the field of chromism and its various types was made.

Keywords: color measurements, thermochromism, smart materials, temperature indicators, pigments, paints and varnishes

Rozważania o termochromizmie należy rozpocząć od zgłębienia zagadnienia barwy. Dopiero poznanie podstawowych praw i zjawisk rządzących światem kolorów, a także wynikających z nich systemów klasyfikacji, pozwala na zrozumienie tej materii. W artykule w zwięzły i przystępny sposób przedstawiono najważniejsze informacje dotyczące postrzegania barw, zwłaszcza w ujęciu matematycznym. Opisano znane i stosowane w sektorze farb i lakierów przestrzenie barw, m.in. CIELab, oraz systemy klasyfikacji kolorów o znaczeniu komercyjnym, takie jak RAL czy Natural Colour System. Opisano metody pomiaru barw oraz przeznaczony do tego sprzęt i odpowiednie normy, których używa się w codziennej pracy technologa wyrobów lakierowych. Dokonano przeglądu stanu wiedzy w zakresie chromizmu oraz różnych jego odmian.

Słowa kluczowe: pomiary barwy, termochromizm, materiały inteligentne, wskaźniki temperatury, pigmenty, farby i lakiery

1. Introduction

Various types of smart materials, including thermochromic pigments, are becoming more and more visible in the global chemical industry market. They are strengthening their position not only because of some innovation but above all, because of their often-unprecedented functionality [1]. All thermochromic materials, by definition, have the ability to change color in response to a change in temperature. After absorbing a certain amount of heat, their crystal or molecular structure undergoes an irreversible or reversible transformation in such a way that it absorbs and emits light of a different wavelength than the initial state. As a result, two different colors can be registered by the observer. There are two key approaches known to obtain pigments and then paint products with thermochromic properties. They are based on the use of liquid crystal and leuco dyes technologies, most often applied in the form of micrometric capsules [2].

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Despite the high attractiveness resulting from the usefulness and decorative properties of thermochromic pigments, currently available industrial solutions are not devoid of some drawbacks and inconveniences. Particularly the high cost of synthesis, difficulties in processing pigments and varnish products obtained with their use, as well as possible or confirmed toxicity, are noteworthy. Due to the above, the use of thermochromic pigments may raise some concerns and prompt reflection on the search for new, environmentally, and user-friendly solutions [3].

It is assumed that in the next few decades, the demand for new thermochromic pigments will increase, especially those of a reversible nature. Industrial areas whose functioning will drive the development of the global market include, among others: the printing, packaging, and textile sectors [4]. Efforts to develop this branch of materials engineering are reflected especially in the paints and varnishes sector. The potential application areas of such materials include the application of solutions dedicated to modern, smart construction and the household appliances sector. Due to the usefulness of thermochromic pigments resulting from their ability to change color, they are usually applied as temperature indicators. This feature allows for the creation kind of heat maps, thanks to which energy losses of residential buildings monitoring and the degree of heating of electrical equipment casings assessment is possible [5].

This article presents the current state of the art of thermochromic materials – pigments and coatings obtained with their use – in the paints and varnishes sector. The article has been divided into two coherent parts. This, the first of them, is devoted to issues related to perception and measurement of color. It is a theoretical introduction enabling a proper understanding of the key property of thermochromic pigments, which is the ability to change color in response to a change in temperature. It covers synthetically and easily described basic issues related to the definition of color, known color spaces, as well as evaluation systems and tools used in industrial practice for color determination. The second one will concern progress in the field of pigments and coatings with thermochromic properties. It will discuss the types of thermochromic materials, especially those of organic origin, and recent developments in the field of thermochromic pigments and coatings based on a review of research published in the last decade. Moreover, a study devoted to market analysis of thermochromic materials will be presented.

The overview of literature and solutions found in the industry was performed with the use of meticulously selected main keywords based on commonly known research databases i.e. Google Scholar and ScienceDirect. Data comes from information brochures or websites of manufacturers and distributors of raw materials for thermochromic paints and varnishes were also used. The cloud containing chosen words connected with the presented subject is shown in Fig. 1. The abovementioned keywords were useful in the process of preparing this work.

Based on the literature analysis on the topics of light-cured coatings on pigments and coatings with thermochromic properties, a graph showing the number of published papers as a function of years between 2013 and 2023 was drawn up (Fig. 2).

Based on the collected numerical data, it can be noticed that the number of papers focused on this topic is increasing from year to year. This increase is quite pronounced for all three keywords

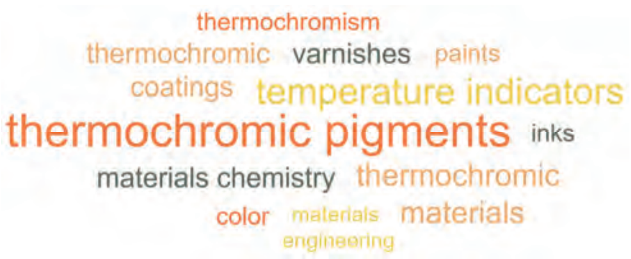

Fig. 1. Thermochromic materials in the paints and varnishes sector – keywords cloud

Rys. 1. Materiały termochromowe w sektorze farb i lakierów – chmura słów kluczowych

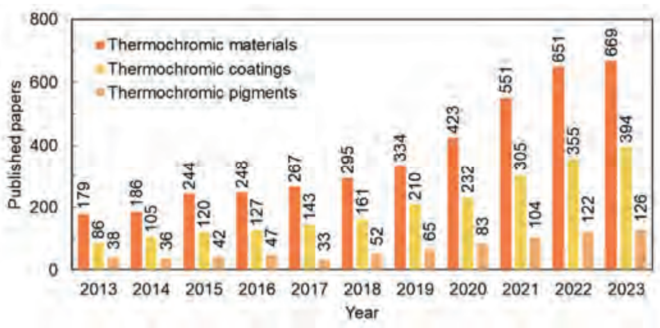

Fig. 2. The number of papers published in the past decade including keywords “thermochromic materials”, “thermochromic coatings”, and “thermochromic pigments”

Rys. 2. Liczba opublikowanych w ostatniej dekadzie prac zawierających słowa kluczowe „materiały termochromowe”, „powłoki termochromowe” oraz „pigmenty termochromowe”

indicated. Due to the visible publication trend, it can be concluded that this trend will continue for at least the next few years. Thus, for the keywords “thermochromic materials”, “thermochromic coatings”, and “thermochromic pigments” in the past decade, the number of published papers has increased from 179 to 669 (+273%), from 86 to 394 (+358%) and from 38 to 126 (+231%), respectively, which correspond to a several-fold increase in the number of published works. The collected data shows that the presented topic is more and more popular and above all very significant. This may be related to the search for new industrial solutions that, apart from being technically useful, would be attractive to the users themselves i.e. non-toxic, more durable and reliable. This situation is similar in the case of the global market of thermochromic materials, especially in terms of pigments. The value of the global thermochromic pigments market in 2022 was between USD 400 and 500 million, and the estimated value of the compound annual growth rate (CAGR) expected for the next few years is approximately 6% [6]. A detailed analysis of the thermochromic pigments market will be presented in the work constituting the second part of this article. Moreover, there is an increasing desire to reduce the negative impact of products from the paint and varnish sector on the natural environment (limiting the use of organic solvents and dyes or pigments with confirmed toxicity to living organisms). This aspect is also a driving force for undertaking new research and analyses.

2. Color and its perception

2.1. Admission word

Color is an issue that can be associated with nearly every existing area of life, from science, through art and industrial design, to cuisine and confectionery. However, it should be remembered that the phenomenon of color has its foundation primarily in physics, and thanks to its laws we are able to make a mathematical description of what the perception of light sensations through the sense of sight allows. By sight, we are able to assess the temperature of an object, which, furthermore, is related to the thermochromic effect. The answer to the question about the definition of color is not easy, since it may be related to both the color impression received by the observer and a physical phenomenon [7].

2.2. Physiological approach

Color perception is a process related to the functioning of the visual system. The process of seeing can be described most generally as follows. The light falling on the cornea of the eye is focused by it, and passing through the pupil and lens reaches the back of the eye, i.e. the retina (Fig. 3). It occupies approximately most of the eyeball and is rich in nerve cells and cells acting as photoreceptors. The latter include: rods – located on the periphery of the retina, responsible for detecting the movement and shape of objects, and cones – collected in the macula of the retina, responsible for detecting color. The blind spot does not have photoreceptors. Its functioning is related to the action of the optic nerve, which when leaving the eyeball, goes straight to the brain. A chemical compound involved in the vision process is rhodopsin. Distributed in the rods, together with other light-sensitive pigments present in the cones of the retina, it is responsible for the generation of a nerve impulse. As a result of light absorption, the chemical structure of rhodopsin undergoes isomerization, inducing the formation of a signaling pathway and a nerve impulse. The resulting images are real images but reduced and inverted. Thanks to the brain’s work, the images are reversed again. Therefore, the color impression depends on the wavelength of the light of the light stimulus affecting our eye. If the beam is not monochromatic, then the visible color will depend on the intensity of all its components. Achromatic colors, which are shades of gray from white to black, and chromatic colors, i.e. all other colors, can be distinguished. Each color can be detected by a healthy organ of vision [8, 9].

2.3. Physical approach

The concept of color is inherently related to light. According to the definition proposed by Zausznica, in physical terms “color is a type of quantitative and qualitative (wavelength, energy, etc.) light radiation, which, as we know, is a form and a limited area of radiant energy (...) acting on and through our eye assessed according to this action” [12, p. 12]. Thus, the visible region of the electromagnetic spectrum covers (depending on the source) a wavelength from about 380 nm to 780 nm, and is limited to infrared and ultraviolet radiation (Fig. 4). To describe light waves, the following are used: wavelength (λ), vibration frequency (ν)

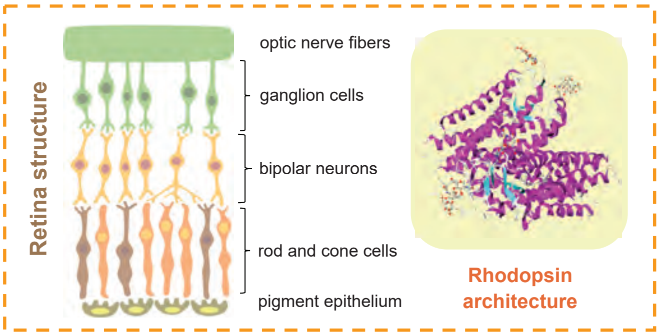

Fig. 3. The structure of the retina with a model of the architecture of rhodopsin consisting of an opsin protein covalently linked to the cofactor 11-*cis*-retinal

Source: own graphics based on [10, 11].

Rys. 3. Struktura siatkówki oka wraz z modelem przestrzennym rodopsyny, składającej się z białka opsyny połączonego kowalencyjnie z kofaktorem 11-*cis*-retinalem

Źródło: opracowanie własne na podstawie [10, 11].

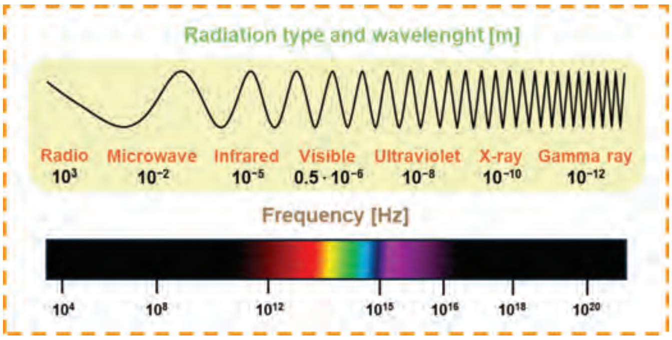

Fig. 4. Diagram of the electromagnetic spectrum

Source: own graphics based on [14].

Rys. 4. Schemat widma elektromagnetycznego

Źródło: opracowanie własne na podstawie [14].

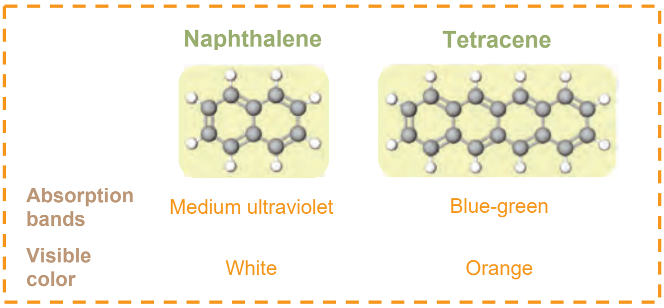

Fig. 5. Naphthalene and tetracene – relationship between the structure, absorption bands, and color of the chemical compound

Source: own graphics based on [16].

Rys. 5. Naftalen i tetracen – zależność między strukturą, pasmami absorpcji a kolorem związku chemicznego

Źródło: opracowanie własne na podstawie [16].

and wave number (v'), the relationships of which are expressed in the equation (where c is the speed of light):

$$\frac{1}{\lambda} = \frac{v}{c} = v'. \qquad (1)$$

The numerical values of these parameters translate directly into the color seen. Today, the wave-particle duality of light is widely known, but the development of science related to light included several theories authored by eminent scientists such as: Newton, Roemer, Huygen, Young, Malus, Fresnel, Foucalt, Maxwell, Hertz, Lorentz and finally Einstein and Planck. The explanation of the wave and corpuscular nature can be found in quantum mechanics and the works of De Broigl, Heisenberg, and Schrödinger [13].

All bodies visible to the naked eye are a source of light. They may be luminous bodies (emitting light) or illuminated transparent/opaque bodies (transforming the light falling on them). Light can transform into reflection, refraction, and splitting, e.g. by obtaining a color spectrum by splitting white light. Passing through any material medium, a beam of light is absorbed. When it overlaps and cancels out with another beam, it undergoes interference, changing the direction of propagation – diffraction, and when the vibrations are arranged – polarization. Any body which temperature is above absolute zero loses energy and emits radiation. The description of the laws of thermal radiation was undertaken by Prevost and Kirchoff, whose achievements were continued by Stefan, Botzman, Wien, and the abovementioned Planck (research on the black body). Subsequent tests were conducted for non-black bodies, i.e. gray and colored bodies. Works on the relationship between the structure of matter and light spectra were the results of Balmer's considerations and research, which were related to the later theories of Rutheford and Bohr [14].

2.4. Chemical approach

In chemical sciences, the concept of color has a wide spectrum and is reflected not only in the processes of monitoring the progress of chemical reactions but above all, it is related to the chemistry of dyes. Such a powerful field of chemistry covers almost every industry: textile, dyeing and varnishing, plastics processing, food, etc. There is also a separate area of chemistry devoted to research on chemical reactions induced by the interaction of electromagnetic radiation – photochemistry, based on the laws of Grothuss, Draper, Hershel, van 't Hoff, and further Einstein and the law of quantum equivalence [15].

It should be remembered that the color of a compound, especially in the sense of substances acting as dyes and pigments, is strictly dependent on its chemical structure and the pattern of absorption bands, which is rarely limited to the visible light region. It is the system of absorption bands of the body in all areas that are responsible for its color. The absorption curves are mainly influenced by the solvents used and the temperature [12].

When analyzing the influence of structure on the color of chemical compounds, it is worth noting that it has been much better understood and described in the case of organic compounds. This is primarily since there is less diversity of atoms in their structure and a certain periodicity, which is manifested, for example, by the presence of a benzene ring and groups formed from the combination of carbon, hydrogen and/or nitrogen (Fig. 5). Theories of the color of organic compounds were initiated by the works of Graebe, Witt, and further by Niezkie, Poraj-Koszyc, Dithley, Wizinger, and Stieglitz. In the classical color theory, it was assumed that the color of a compound depends on the grouping of atoms in its molecule (chromophore), and the compound becomes a dye only after introducing further groups into the molecule (auxochrome). Later in the years, electron theory was developed (works by Sklar and Förster). According to its content,

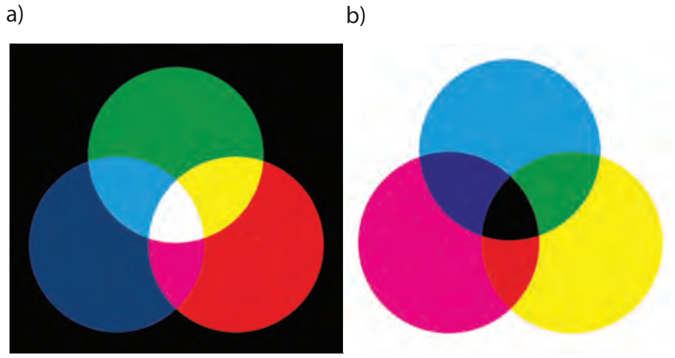

Fig. 6. Color mixing: a) additive method, b) subtractive method

Source: own graphics based on [19].

Rys. 6. Mieszanie barw: a) metoda addytywna, b) metoda subtraktywna

Źródło: opracowanie własne na podstawie [19].

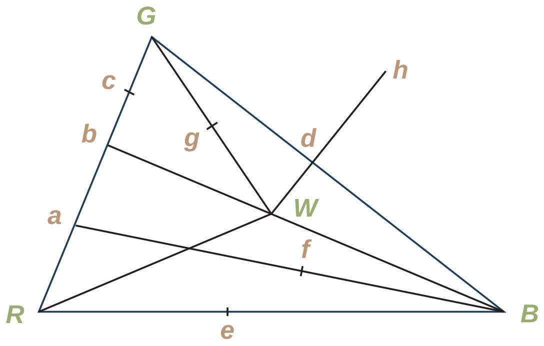

Fig. 7. Color triangle (RGB – primary colors, W – white point)

Source: own graphics based on [12].

Rys. 7. Trójkąt barw (RGB – barwy podstawowe, W – punkt bieli)

Źródło: opracowanie własne na podstawie [12].

the color of an organic compound is the result of the presence of conjugated π electron systems in its molecule. The systems can be easily excited, thereby absorbing radiation of a specific wavelength in the visible light range, while the rest are reflected, resulting in the perception of a specific color temperature [12].

3. Mathematical approach – foundations of colorimetry

3.1. Admission word

The light reaching the eye may have different spectral composition and intensity. If the difference in beams results from variable intensity, we talk about brightness contrast (quantitative difference – photosensitivity). However, if it depends on the spectral composition, we are dealing with color contrast (qualitative difference – color sensitivity). The science of color is based on Newton's experiments on the splitting of white light on a prism and the isolation of primary colors, i.e. red, yellow, and blue, which create the first model of the RYB color space (from the first letters of colors) [17]. The foundations for the concept of color space are, in turn, Young's postulates, which state that there are three types of cells (eye cones) that are sensitive to visible light of a specific range. The development of these assumptions was the formulation of the Young-Helmholtz theory about three types of cones in the eye related to the reception of blue, green, and red light [18].

Monochromatic beams always create the same color impression. However, when more than one beam is directed at the organ of vision, it is possible to perceive an infinite number of combinations (pairs and triads) that can produce the same color impression. Therefore, two colors or their groups that beams, when added, create white are called complementary colors [19].

One of the attributes of color is its brightness, which organizes the position of chromatic and achromatic colors between black and white. The second attribute of a color is its hue, i.e. a feature that allows two colors to be distinguished as different. The next and last attribute is color saturation, which means that a color comes close to white without changing its hue (the difference between chromatic colors with the same hue and brightness). The change in chromatic color from white is called the degree of saturation, and to white, the degree of unsaturation. When referring to black, we are talking about the degree of color purity. All the color attributes mentioned above are examples of qualitative contrast. The creation of colors may be the result of their additive mixing (both light, paints and varnishes), which is governed by Grassmann's laws. The first one concerns trichromaticity (each color can be expressed using three independent ones), the second one deals with continuity (each change in the color of one of the components of the mixture causes a change in the mixed color), and the third one is devoted to additivity (the color of the mixture does not depend on the spectral composition but on the color of the ingredients). These laws constitute the foundation of the science of color and are analogized in the arithmetic (vector) approach. Another case of color mixing is subtractive synthesis, in which the added colors become closer to black and lose their intensity (Fig. 6) [19].

To present the color on a chart, the so-called color triangle (Fig. 7), consisting of three primary colors and a white point was proposed. By combining them, a color that can be represented as a point located inside the color triangle is obtained. This, in turn, is the basis for the classification of color models and their spaces [12].

3.2. Determination of color in space

When talking about defining color, you should use the terms "model" and "space". And what is the difference between these two? It should be remembered that a color model is its representation in numerical form (mathematical/informatic description), which is subject to mapping in the color space. The color space allows for the reproduction of real colors in the color range (it defines and organizes the range of colors that can be obtained/produced). Color spaces include visible radiation spectra (as mentioned, λ from 380 nm to 780 nm), which mathematical models are represented in a three-dimensional color system. Existing color models are useful because they are characterized by varying degrees of similarity to color perception with the vision organ. The diversity of color spaces is due to the fact that they can be represented by physically existing color samples or refer to mathematical structures. Colors in space are defined in relation to accepted standards, which are most often CIELab or CIEXYZ color spaces. This is due to the fact that these spaces are designed to include all the colors seen by the average observer. All the most important color models and color spaces are included in the content of specialized standards, the application area of which covers a number of industries, not only the paint and varnish sector [20].

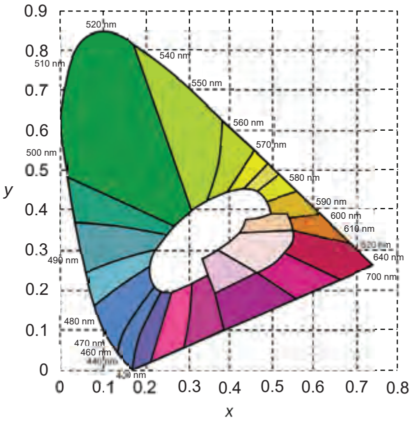

Fig. 8. Chromaticity diagram

Source: own graphics based on [23].

Rys. 8. Wykres chromatyczności

Źródło: opracowanie własne na podstawie [23].

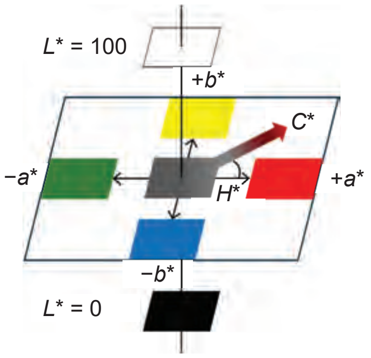

Fig. 9. CIELab color space diagram

Source: own graphics based on [24].

Rys. 9. Model przestrzeni barw CIELab

Źródło: opracowanie własne na podstawie [24].

3.3. Color space models

A detailed description of the known color space models would be difficult, primarily due to the considerable volume of the presented content. For this reason, some of the color spaces will be briefly described, and more attention will be devoted to the spaces from the CIE family, especially CIELab, which, to the best of the authors' knowledge, is the space most often encountered in the professional work of coating technology specialists. Thus, in the classification of color space models, the following examples should be distinguished [21]:

- RGB – a model which name is an acronym of the English names of the colors included in it, i.e. R (red), G (green), and B (blue). The principle of its operation is based on creating a new color (derivative) by mixing three base colors in certain proportions. This model is a theoretical model that includes additive mixing methods. Used in analog and digital techniques, especially in computer science. Further color space models were created on this basis, such as sRGB, scRGB, Adobe RGB, ProPhoto RGB, Adobe Wide Gamut RGB or DCI-P3.
- CMYK – a model which name is an acronym of the English names of the colors included in it, i.e. C (cyan), M (magenta), Y (yellow) and K (black/key color). At the same time, the same term covers a set of basic printing ink colors with similar colors. Therefore, the most important application area of such a model includes not only computer graphics, but also printing. According to this model, new colors are created by superimposing primary colors in specific proportions, not by mixing them. Hence, the obtained color can be up to 400% of the basic colors. Its form that excludes the black color is the model CMY.
- HSV (or HSB) – model, taking the form of a cone based on the color wheel, the name of which is an acronym of the English-language names of the color description components, i.e. H (hue) as an angle on the color wheel from 0° to 360°, S (saturation) as the radius of the cone and V (value) or B (brightness) as its height. According to the assumptions of this model, the beginning of each color is white, the spectrum of which is partially absorbed or reflected. Intermediate colors are distributed between red (0° or 360°), green (120°) and blue (240°).
- HSL – model, taking the form of a cone based on the color wheel, the name of which is an acronym of the English-language names of the components of the color description, i.e. H (hue) with values up to 359°, S (saturation) and L (lightness), both in the ranges from 0 to 1 or from 0% up to 100%. According to its assumptions, it is possible to assign a point to each color perceived by the observer in the space of its components. Its creation is historically linked to the advent of color television. The third color space model related to the HSV and HSL models is the HSI model, in which the letter I stands for intensity.
- YUV, YIQ, YDbDr, YPbPr – color space models used in analog television systems, replaced by the YCbCr model in digital systems. Popular especially during the transition from black and white to color receivers. In the above models, Y stands for luminance, and UV, IQ, DbDr, and PbPr are related to chrominance.

3.4. CIE color spaces

The CIE family of color spaces originated in the first half of the 20th century, when the International Commission on Illumination proposed the creation of the CIEXYZ space, also known as CIE1931. To this day, it is considered a standard that gave rise to subsequent color spaces created on its basis, i.e. CIELUV and CIELab. The abbreviation CIE comes from the French name of the commission, i.e. Comission Internationale de l'Eclairage. Determining the color in this three-dimensional space is based on the numerical data of the tristimulus coordinates X and Z, which correspond to the percentage of RGB colors, and Y means luminance. Color reproduction in this system is possible thanks to the use of a color triangle, i.e. a chromaticity chart. As mentioned earlier, the idea behind the creation of CIEXYZ was the need to capture all the colors that could be observed by an untrained viewer. In fact, the CIEXYZ color space from 1931 is widely used to this day. However, due to a certain imperfection of the proposed system related to the uniformity of color perception, efforts were made to develop it and create new, improved color spaces [22].

The best tool used to illustrate how the eye will select light in the analyzed spectrum is the chromaticity diagrams (Fig. 8). The diagram area shows the range of colors that make up the shape of the tongue. The bent part is the so-called spectral locus, which corresponds to a monochromatic beam of light with a specific color and shade. The lower edge is called the line of purples, for colors that do not have a monochromatic counterpart. All colors that exist between two points on the diagram can be obtained by mixing the starting colors. In the case of three colors, the resultant color will always lie inside the triangle formed by their points [23].

Nearly 45 years later, the CIELab color space, also known as CIE 1976, was introduced, which was a modification of the Lab Hunter space. The creation of this method was correlated with the above-mentioned unevenness of color perception. Through an appropriate mathematical transformation of the CIEXYZ space, the new CIELab space was to be free of this snag and thus constitute the first uniform color space. Fig. 9 shows a diagram graphically presenting the CIELab color space [24].

In principle, all colors separated by the same distance, i.e. color difference *ΔE*, should be perceived by the observer as identically different. In reality, however, even in this case we are dealing with a certain unevenness and discrepancy between the actual color perception and the *ΔE* values. In the industrial practice in paints and varnish technology, it is assumed that two colors will be distinguished by an untrained observer as two different ones when $2 < \Delta E < 3.5$, and the observer gains complete color perception confidence when $\Delta E > 5$. In this color space model, color is described by three components, which are parameters: L^* – brightness (from black to white), a^* – color from green to red, and b^* – color from blue to yellow. There are also parameters C^* – chroma and H^* – hue, for which the difference Δ can also be determined. The transformation of the CIEXYZ space into CIELab occurs according to the following equations [22]:

$$L^* = 116 \cdot Y^* - 16, \tag{2}$$

$$a^* = 500 \cdot (X^* - Y^*), \tag{3}$$

$$b^* = 200 \cdot (Y^* - Z^*), \tag{4}$$

where $X^* = \sqrt[3]{\frac{X}{X_n}}$

for $\frac{X}{X_n} > 0.008856$

or $X^* = 7.787 \cdot \left(\frac{X}{X_n}\right) + 0.138$

for $\frac{X}{X_n} \leq 0.008856.$

Calculation of Y^* and Z^* in analog to X^*, where X_n, Y_n and Z_n are the tristimulus values of a perfect white diffuser at the chosen CIE standard illuminant (A/10° or D_{65}/10°), and then [22]:

$$C^*_{ab} = \sqrt{a^{*2} + b^{*2}}\ , \tag{5}$$

$$h^*_{ab} = \arctan \frac{b^*}{a^*}, \tag{6}$$

$$\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}\ . \tag{7}$$

Another color space belonging to the CIE family is the CIELUV space. Its creation is related to the search for a model that would allow for the linearization of the perception of color difference, i.e. enabling the creation of the so-called perceptual uniformity. This treatment allows to achieve the effect of a smooth, almost smooth transition between individual colors. CIELUV is a development of the CIEXYZ and CIEUVW models from 1964, which, like the CIELab space, was introduced in 1976. Colors classified according to the CIELUV model are described in Adams chromatic valence color space. The cylindrical version of the CIELUV space is known as CIELCh(uv), and the CIELab model is known as CIELch(ab). These use polar coordinates instead of Cartesian coordinates, where: *L* – lightness, *C* – chroma, *h* – hue angle [21, 25].

In the professional literature and industrial practice, some inaccuracies can be noticed regarding the nomenclature of the above spaces, which are often unified by changing the order of symbols or reducing various terms to one, i.e. LCh, LCH, LCh(ab), LCH(ab), HLC, and further with LCh(uv). This procedure is a great simplification and may mislead the recipient.

3.5. Commercial color classification systems

Considerations about color classification systems originate from Munsell's work. The organization system he proposed was undoubtedly the starting point for all contemporary color systems. The foundation of Munsell's approach is the organization of colors based on: hue (basic color), value (lightness), and chroma (color intensity). Created in the first years of the 20th century, the study was initially used only to measure and classify soil color, only later it was applied in other areas. The innovation of this solution consisted of the separation of color parameters into separate, perceptually uniform dimensions, and then in the presentation of colors in a systematic three-dimensional space. Although nowadays the Munsell system has been partially replaced by newer classification systems, e.g. CIELab, due to its specificity based on aesthetic experiences (visual reactions), it is still widely used. One of the consequences of the creation of the Munsell color classification system was the development of color charts. Color charts are a useful tool in color management work. They enable comparison of individual colors, e.g. in calibration processes, and quality assessment, e.g. ColorChecker or Shirley cards, and, above all, color selection. It is color selection charts that are most important in the industrial practice of the paints and varnishes sector [26–28].

3.6. RAL color standard

The RAL color standard is the key commercial color classification system from the perspective of a coating technology specialist. The idea behind its operation is to use standards against which the developed color is compared. Created by the Reichsausschuss für Lieferbedingungen in the first half of the 20th century, it initially included only 30 colors. Today, 2,016 colors stand out for the basic RAL Classic system. The RAL Design system has been expanded to 1,825 colors, and the RAL Effect system has 490 colors. RAL solutions include all types of organic coatings applied using any techniques, e.g. hydrodynamic and pneumatic spraying or powder coating, applied in numerous sectors of industry and fine arts. This system is not assigned to specific paint and varnish producers, which is why it is so eagerly used all over the world. Colors organized by the system are marked as follows [26–28]:

- RAL Classic – coding using four-digit numbers, where the first two digits mean respectively shades of: 10 – yellow, 20 – orange, 30 – red and pink, 40 – purple and violet, 50 – blue, 60 – green, 70 – gray, 80 – brown, 90 – white and black. A traditional system, used since the beginning of RAL. Example: RAL 6003.

Fig. 10. Color charts of NCS and RAL systems

Rys. 10. Wzorniki kolorów systemów NCS oraz RAL

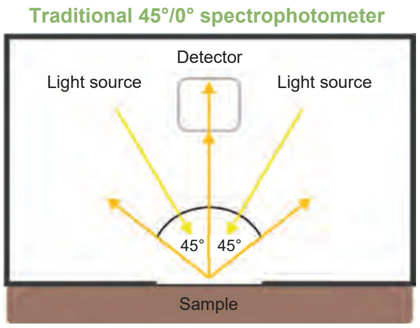

Fig. 11. Construction diagram and principles of operation of traditional spectrophotometer

Source: own graphics based on [33].

Rys. 11. Schemat budowy i zasada działania tradycyjnego spektrofotometru

Źródło: opracowanie własne na podstawie [33].

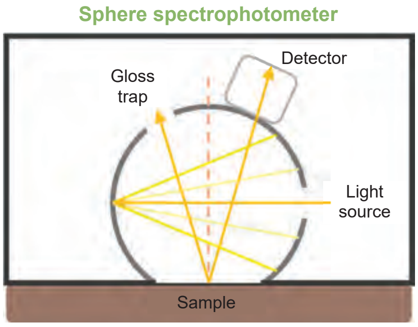

Fig. 12. Construction diagram and principles of operation of sphere spectrophotometers

Source: own graphics based on [33].

Rys. 12. Schemat budowy i zasada działania spektrofotometru sferycznego

Źródło: opracowanie własne na podstawie [33].

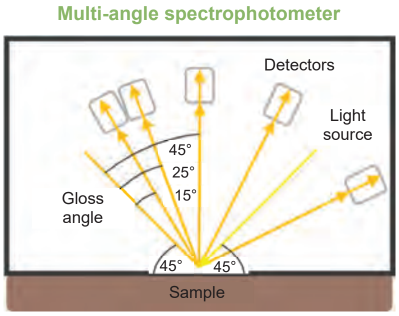

Fig. 13. Construction diagram and principles of operation of multi-angle spectrophotometer

Source: own graphics based on [33].

Rys. 13. Schemat budowy i zasada działania spektrofotometru wielokątowego

Źródło: opracowanie własne na podstawie [33].

- RAL Design – coding using seven-digit numbers, where the first three digits correspond to the position of the shade on the color circle, the next two digits describe the brightness (where 0 is black and 100 is white), and the last two the level of colorfulness (where 0 is gray and 100 is full color). Example: RAL 200 30 30.
- RAL Effect – coding using four-digit numbers based on the classification of color families, which are marked with three-digit numbers. After the hyphen, the fourth digit corresponding to the tone and metallic shades is written. Example: RAL 530-6.

3.7. Natural Colour System

The idea of the Natural Color System (NCS) is based on the perceptual vision of colors, not on the effect of mixing colors. This system was developed based on the research and publications of Edwald Herning, whose origins date back to the end of the 19th century. The currently known version of this system was developed in 1964 by the Swedish Color Center Foundation, and its operating principle was illustrated in a dedicated atlas. Nowadays, the NCS color classification system is one of the most popular tools for systematizing colors. The areas where it is particularly popular are Scandinavian countries and Spain. The use of NCS does not involve any limitations resulting from the physical characteristics of the element to be classified. The color notation is clear and is based on the markings below. This NCS system distinguishes six basic colors: white (W), black (S), yellow (Y), red (R), blue (B), and green (G), which create three opposing pairs: W-S, G-R and Y-B. Colors are defined by three main values, expressed as %: degree of blackness (*s*), chromaticness (*c*), creating the so-called nuance, and hue (*h*). The combination of *s* and *c* add up to less than or equal to 100%, the rest is whitening (*w*). The NCS color space is illustrated in a three-dimensional double-cone model. Therefore, in simple terms, it is presented in two sections, i.e. a color circle (a kind of color wheel with four evenly spaced primary colors) and a shade triangle (enabling you to find a shade based on % *s* and *c*). Notation for the example color S 2050-B50G: S – NCS catalog edition, 2050 – nuance (20% *s*, 50% *c*, 30% *w*), B50G – hue, 50% G and 50% B, i.e. blue with 50% green. A photo showing an example of the color charts of NCS and RAL systems is shown in Fig. 10 [26–28].

3.8. Pantone Matching System

Another commercially used color classification system is the Pantone Matching System (PMS). The method for color organization proposed by Pantone LLC was developed based on the existence of 18 pigments, from the mixing of which a complete color palette is obtained (the basic palette includes 1,761 colors). Due to the use of white and black pigments, the transfer of PMS colors to the RGB and CMYK scales was not fully possible for a long time. It was only in 2007 that the Pantone Goe System was developed, eliminating this inconvenience. In PMS, colors are marked with three- or four-digit numbers with the letter C (coated/glossy paper) or U (uncoated paper). A small part of the colors are named according to the actual color, e.g. Pantone Blue Process C. There is also a whole range of colors with a decorative effect, such as Pantone Metallics or Pantone Pastels & Neons. The PMS application area includes primarily printing solutions, but also all branches of industry and art belonging to the so-called creative industries and related to graphic design, such as industrial or clothing design [26–28].

4. Colorimetric measurements

4.1. Admission word

Due to the difficulties in creating a unified color representation system, there was a need to express it mathematically. The development of spectrophotometry came to the rescue, thanks to which spectrophotometers were invented – devices used for the numerical expression of color [29]. The principle of working is to expose the tested material to radiation and measure the amount of light reflected or transmitted by the sample in different parts of the spectrum. First, a beam of light is directed at the sample, which – depending on the geometry of the spectrophotometer – is split in the appropriate directions and then hits sensors that measure the amount of incident light. Each sensor is in a fixed position and can measure the appropriate part of the spectrum. The resulting signals then go to a detector, which transforms them into a set of numbers that create a spectrum – a spectral curve – also known as a spectral fingerprint [30].

In industrial practice, spectrophotometers are the basic device used for color measurements. An undoubted advantage is the ability to measure a wide range of products, from liquids through plastics, paper, metal, and even fabrics. There are three types of spectrophotometers commonly applied in colorimetric measurements, and the choice of the appropriate one depends on the individual needs of the user [31].

4.2. Traditional spectrophotometer (0°/45° or 45°/0°)

In the case of traditional 0°/45° spectrophotometers (Fig. 11), the light beam is directed at the tested sample at an angle of 45°, and the detector receives the light signal reflected perpendicular to its surface, at an angle of 0°. It is considered one of the most popular spectrophotometers and is used primarily for measurements on smooth or matte surfaces [32].

4.3. Sphere spectrophotometer (diffuse/8°)

Measurements carried out using sphere spectrophotometers (Fig. 12) use the so-called sphere geometry. The tested material is illuminated from all directions so that the light is scattered, and the detector collects signals reflected at an angle of 8°. Additionally, the device is equipped with low gloss and highly reflective material, thanks to which almost 99% of the incident light is effectively reflected, allowing the appropriate beams to reach the detector. This type of spectrophotometer is suitable for measurements on high-gloss surfaces (e.g. shiny, mirror) [34].

4.4. Multi-angle spectrophotometer

Multi-angle spectrophotometers (Fig. 13) provide significantly extended possibilities for color observation. Their use provides the possibility to determine the color of the sample from different angles. In this case, the beam falls on the sample at an angle of 45°, and the spectrum is recorded by several detectors set at angles of 15°, 25°, 45°, 75° and 110°. So far, such devices are used primarily in the automotive and cosmetics industries. Observation of color from various angles allows to examine materials covered with metallic, pearl, or mica pigments that give the surfaces special effects [35].

4.5. Colorimeter

Another useful device for determining color is a colorimeter. In this case, color measurement is not as precise as in the case of tests using spectrophotometers. Colorimeters imitate the way the human eye perceives color. The light beam falling on the sample, after being reflected towards the detector, passes through the red, green, and blue filters. Thanks to this, basic chromatic values (RGB) are extracted, which correspond to how the human eye reacts to light and color. The use of colorimeters is often limited to basic color assessment, mainly in cases where restrictive quality control is not required [36].

4.6. Densitometer

Apart from the spectrophotometers and colorimeters discussed above, densitometers are also useful in color measurements. They are used to measure the optical density of transparent and opaque materials, especially when working in the CMYK color model. Mainly used in the printing industry to measure, among others, ink density and print thickness. There are two types of densitometers: transmissive (useful for measuring light transmittance through the tested material) and reflective (used for measuring the percentage of light reflected by the exposed material) [37].

4.7. Colorimetry in industrial practice

Having basic knowledge about the structure and principles of operation of color measurement devices, the practical work can be started. In the paints and varnishes sector, the field of colorimetry seems to be one of the key issues determining not only the quality of finished products, but above all, the aesthetic feelings of the recipient and, subsequently, favorable sales results. The main manufacturers of devices used for colorimetric measurements, not only in the paint and varnish sector, include X-Rite, HunterLab, Konica Minolta, and 3nh. The solutions they offer include a wide range of products, from spectrophotometers, through colorimeters, to densitometers, without which colorimetric quality control of coatings would be impossible. Color measurements, both those performed during laboratory tests and on an industrial scale, are carried out in accordance with the rules presented in dedicated standards. Thus, depending on the customer's expectations, the type of product being tested, its purpose, and geographical area, there are several most important standards for the paints and varnishes sector. A list of them, including numbering, title, and short description, is presented in Table 1.

5. Chromism and colorimetry in scientific reports

Chromism is a phenomenon involving a reversible change in the color of a substance under the influence of physical or chemical environmental conditions. When an external factor affects the tested substance, its electronic, conformational, crystal, or

Table 1. List of key colorimetric standards used in the paints and varnishes sector
Tabela 1. Wykaz najważniejszych norm kolorymetrycznych stosowanych w sektorze farb i lakierów

Number	Title	Short description	Ref.
ISO/CIE 11664	Colorimetry Part 1: CIE Standard Colorimetric Observers Part 2: CIE Standard Illuminants Part 3: CIE Tristimulus Values Part 4: CIE 1976 *L*a*b** Colour Space Part 5: CIE 1976 *L*u*v** Colour Space and *u′*, *v′* Uniform Chromaticity Scale Diagram Part 6: CIEDE2000 Colour-Difference Formula	These documents specify: colour-matching functions for use in colorimetry, define three CIE standard illuminants for use in colorimetry, methods of calculating the tristimulus values of colour stimuli, method of calculating the coordinates of the CIE 1976 *L*a*b** and CIE 1976 *L*u*v** colour space and the method of calculating colour differences according to the CIEDE2000 formula	[38]
DIN 53236	Colouring materials – Conditions of Measurement and Evaluation for the Determination of Colour Differences for Paint Coatings, Similar Coatings, and Plastics	This standard specifies: measurement and assessment conditions for determining colour differences in paints, varnishes, and similar coatings and plastics, especially with regard to the determination and assessment of color changes subjected to physical or chemical stress	[39]
SAE J1545	Instrumental Color Difference Measurement for Exterior Finishes, Textiles, and Colored Trim	This standard specifies: practice applies to parts and materials used in vehicle manufacture which are intended to be acceptable color matches to a specified color standard, used for non-transparent materials such as topcoat paint finishes, interior soft trim, interior and exterior hard trim, and exterior film	[40]
VW 50195	Colorimetric Evaluation of Exterior Automotive Paint Finishes	This standard specifies: evaluation criteria for evaluating color deviations and the measurement and evaluation conditions for the device-based determination of color differences for paints in body color	[41]
PN-ISO 7724	Paints and Varnishes – Colorimetry Part 1: Principles Part 2: Colour Measurement Part 3: Calculation of Colour Differences	These documents specify: colorimetric terms and basic requirements and a method for determining the color coordinates of paint coatings and related materials and a method for quantitative colorimetric assessment of small color differences between paint coatings	[42]

physical structure can undergo transition, which is manifested by a change in color [43]. Depending on the factor causing these changes, many types of chromism can be mentioned, and from the point of view of up-to-date scientific research, those that deserve special attention are [44]:

- thermochromism – color change occurs as a result of temperature change,
- halochromism – a color change occurs as a result of a change in pH,
- photochromism – the color change is caused by light radiation,
- electrochromism – the color change is caused by an electric current,
- magnetochromism – the color change is caused by a magnetic field,
- solvatochromism – color change due to the polarity of the solvent.

The chromism effect can be easily monitored using colorimetric measurements, and due to their immense application potential, they are discussed in many fields of science. Most of the published works base the investigation of color changes on the classic measurements using CIELab color space, sometimes combined with UV-Vis spectroscopy [45]. Applications of chromic materials often discussed in the scientific literature include i.a. smart textiles [46, 47], temperature indicators for the food industry [48, 49], smart printing inks [50], biosensors [51], or gas detectors [52].

Pu and Fang [53] have proposed studies on the thermochromic response of leuco dye-based microcapsules working at refrigeration temperatures (from 1°C to 8°C). For this purpose, crystal violet lactone and lauryl gallate were encapsulated via coacervation with the use of methylcellulose. The effect of the mass ratio of these three components on the intensity of color change was evaluated via color measurements using CIELab chromaticity indexes. The best thermochromic response was recorded for the system where crystal violet lactone, lauryl gallate, and methylcellulose were mixed in a 1 : 3 : 125 wt% ratio. Gradual color changes from deep blue to white were observed at temperatures from 1°C to 8°C. The authors state that such a system could have a potential as low-temperature reversible temperature indicators, especially in the food and packaging industries – for example for the monitoring of food freshness.

Another research on the color change in thermochromic leuco dye systems was proposed by Šťaffová et al. [4]. The group was working on the development of polyester-based textiles dyed with red-colored leuco dyes dispersion in polyurethane. The activation temperatures of investigated dyes were stated at 38°C, 50°C, and 60°C. The colorimetric measurements were the basis of reported research and investigation with the use of two different spectrophotometers was conducted. The first one was applied for the light stability tests, whereas the second was for the temperature-dependent analysis of prepared textiles. The colorization and decolorization effect was investigated and the results were compared to the thermal properties of the pigment. The thermochromic nature of pigments was confirmed by determining the phase change transitions and dynamic color change temperatures. However, the photostability was not sufficient enough, and thus, an attempt for its improvement was made by adding UV and HALS stabilizers into the dyes dispersion. Such a solution can be a step towards the development of novel smart textiles, which is a promising field in materials technology.

Panák et al. [54] have presented research on a thermochromic leuco dye-based ink system containing crystal violet lactone, bisphenol A, and 1-tetradecanol. A dynamic color change influenced by thermal treatment was investigated with the use of a digital camera. The changes in the color of samples in different forms: bulk system, dry pigment, water- and solvent-based ink formulations, and their dry layers coated on different paper substrates were analyzed via color measurements in CIELab color space. The water-based ink coated on paper substrates exhibited higher values of color change than the layers of solvent-based ink. It was shown that the water-based coatings had much higher chroma and were less influenced by the paper substrate. The resulting system was investigated for the potential use as smart printing inks suitable in i.a. textile production.

An attempt at a smart packaging system was made by Andretta et al. [55]. The group has prepared biopolymeric thin films based on natural colorants with halochromic properties. The study was based on the development of starch-based thin films by thermocompression with blueberry (*Vaccinium corymbosum* L.) residues as a pH change indicator. Analysis of color change was done using classic colorimeter and chromatic values were determined in CIELab color space. Thanks to the anthocyanins content in blueberry residue, samples exhibited visually perceptible color changes from pink to yellow-brownish in the pH range between 2 and 12. The analysis confirmed the potential of the anthocyanins as color indicators, which could be useful in i.a. the food packaging industry.

One of the interesting paths for halochromic dyes application is gas sensors. Park et al. [56] have described the preparation of pH-sensitive textiles with organic dyes (such as e.g. Rhodamine B) with the ability to NH_3 and HCl gas detection. The dyes were incorporated into polyester fabrics using the screen-printing method. The colorimetric measurements were performed using a traditional 0°/45° spectrophotometer. High detection rates for obtained sensors were stated and visible color changes under alkaline or acidic conditions, even at low gas concentrations were confirmed. What's more, great durability and reversibility after washing and drying were shown which allows the fabricated textiles to be further applied in gas sensing solutions.

Chromic effects can also be used in the field of photoluminescent technologies. Al-Qahtani et al. [57] reported research on photochromic transparent wooden smart windows with color-switching ability in the UV and visible spectrum regions. Here, lignin-modified wood substrates with a methyl methacrylate and photoluminescent lanthanide-doped aluminum strontium oxide pigment were investigated. A translucent wooden substrate characterized by great photo- and thermal stability was obtained. A color change from colorless in visible light to green under irradiation with UV light was determined by CIELab colorimetric measurements. It was noted that the discussed transparent luminescent woods show significantly improved UV protection and fast reversible photochromic response.

Another example of materials exhibiting chromism are electrochromic devices which are particularly applied in optoelectronics, for example in electronics-free, self-powered sensors. Aller-Pellitero et al. [58] were working on a glucose biosensor connected to a Prussian blue electrode for blood testing and diabetes screening. In this work, conventional transparent electrodes were replaced with a screen-printed layer of semi-conducting antimony tin-oxide particles coated with Prussian blue. Thanks to the electrochromic response of applied dye, it was possible to observe color changes in the sensor system even with the naked eye.

Biosensing applications have attracted researchers for many years now. One of the routes to fabricate such systems is the employment of solvatochromic materials. Bisphenol-based optical sensor with the ability to change color under the influence of different solvents was developed by Kakoti et al. [59]. Here, a sensor for the detection of water, heavy water, and hydrogen peroxide was developed. The bisphenol-based optical sensor 4,40-((4-hydroxyphenyl)methylene)bis(2,6-dimethyl phenol) was synthesized in a reaction of 4-hydroxybenzyladehyde and 2,6-dimethyl phenol. The solvatochromic effect was confirmed in different organic solvents (i.a. ethanol, ethyl acetate, acetonitrile, acetone). The color measurement results expressed as RGB coordinates in different solvents varied from light yellow, through orange to deep green. Moreover, it was shown that the efficiency of colorimetric response was improved through the deprotonation of the bisphenol system.

For some applications, one possible chromic response can be not efficient enough. Thus, there are also attempts at the fabrication of multifunctional chromic materials. Atav, Ergünay, and Akkuş [60] proposed research on the production of smart cotton-based sensor textiles by applying a combination of thermochromic, halochromic, and photochromic dyes. Such a system was designed to provide a few functions such as information on whether the ambient temperature is risky for the worker (thermochromic), detection of dangerous chemical vapor in the work environment (halochromic), and creation of a visual effect for the protection against UV radiation (photochromic). A thorough analysis of the color changes via measurements in CIELab color space was performed. It was shown that the prepared chromic fabrics have a good reaction to the changes in pH, temperature, and light. Moreover, samples maintained the long-lasting ability to color changes – even after 20 cycles of measurements.

6. Summary

The progress of materials engineering is driven by newer and more advanced discoveries in the fields of materials chemistry and physics. One of the milestones of this discipline is undoubtedly intelligent materials. Due to several unique properties, including the ability to change color in response to temperature changes, they have given rise to a new chapter in materials science.

When talking about thermochromism, all considerations should begin with exploring the issue of color. Therefore, this matter is the subject of this work. An attempt was made to present the knowledge gathered logically and coherently. Various approaches to color perception are described, focusing on the mathematical approach. It is this that lays the foundations for modern color systematics and colorimetry in general. Starting from the color space and ending with commercial color classification systems. Color measurement methods along with dedicated equipment and standards were also presented. At last, a brief review of the state of knowledge in the field of chromism was reported.

The above article is the first of two parts of a larger review devoted to thermochromic materials in the paints and varnishes sector. The authors' intention was to introduce the reader to the world of colors to fully understand the phenomenon of thermochromism as well as the latest achievements in the synthesis or application of pigments, and further thermochromic coatings to which the second part of the article will be devoted. Both parts of the work, this and the next one, were compiled from the perspective of practitioners – coatings technology specialist. However, they are addressed to a much wider audience, and that is all those for whom the issue of color and thermochromism is a topic worth knowing.

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CRediT authorship contribution statement
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