Colorant Materials, Additives in Plastic **Technology**

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Abstract

Pigments and dyes are coloring substances, i.e., substances imparting a specific color to the substrate being colored (Carolin et al. 2015). Pigments and dyes are distinctly different types of colorants. A pigment is a finely divided solid which is essentially insoluble in its polymeric application medium. Pigments are incorporated by a dispersion process into the polymer, while it is in a liquid phase, and after the polymer solidifies, the dispersed pigment particles are retained physically within the solid polymer matrix. In contrast, a dye dissolves in the polymeric application medium and is usually retained as a result of an affinity between individual dye molecules and polymer molecules. Pigments are generally preferred to dyes for the coloration of plastics mainly because of their superior fastness properties, especially migration resistance (Maile et al. 2005). Pigments may be introduced into plastics by a variety of methods. Direct dry coloring, in which the pigment is incorporated into the molten polymer often along with other additives using high-shear dispersing equipment, may be used. However, many manufacturers of plastic articles find it more convenient to make use of pre-dispersed concentrates or master batches of pigment in a liquid additive such as a plasticizer or in a compatible resin. Such concentrates are then easily incorporated by mixing into the final polymer composition at an appropriate stage of the processing sequence. Pigments are conveniently classified as either inorganic or organic types. The properties of a pigment are primarily dependent on its chemical constitution. However, other factors influence the properties as a result of the fact that pigments are used as solid crystalline particles.

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 \oslash Springer-Verlag GmbH Germany 2017

S. Palsule (ed), Polymers and Polymeric Composites: A Reference Series, https://doi.org/10.1007/978-3-642-37179-0_81-1

Keywords

Pigments • Dyes • Colorants • Dispersing agents • Paint • Solvent

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Introduction

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Organic pigments generally show an increase in color strength as the particle size is reduced, while with many inorganic pigments, there is an optimum particle size at which the color strength reaches a maximum. Other important factors which influence the dispersion properties in particular are the degree of aggregation of pigment particles and the nature of the particle surfaces (Kaminska [2005\)](#page-15-0).

Colored polymers with excellent pigmentary properties were obtained by copolymerization of pigment moieties of the naphthol AS series which contain reactive polymerizable groups for copolymerization with other unsaturated vinyl monomers such as acrylamide, acrylonitrile, styrene, and methyl methacrylate. The polymeric compounds showed improved chemical resistance to common solvents encountered in surface coating applications. Preliminary investigation showed that these compounds have good potential as raw materials for the paint industry since they have good affinities to substrates and superior light fastness when used for the pigmentation of paints (Achi and Myina [2013\)](#page-15-0).

Pigments

Pigments serve three main functions: the optical function of providing color, opacity, and gloss, a protective function with regard to the surface underneath the paint and with regard to the binder which can be destroyed by UV, and a reinforcing function for the paint itself in that they help the binder to stick. Pigments are composed of tiny solid particles less than 1 μm in diameter, a size that enables them to refract light (light has wavelengths between 0.4 and 0.7 μ m).

For the pigment to be effective, it has to be evenly dispersed throughout the solvent and in contact with the solvent. Surrounding pigment particles is a layer of moist air and, in some cases, other gases. To bring the pigment into contact with the solvent, this layer has to be displaced, and this displacement is known as wetting. If a pigment is not properly wetted in paint, it may result in color streakiness in the finished paints; thus solvents and pigments must be chosen that result in a wellwetted pigment. Wetting and dispersing agents are used to improve the wetting properties of the resin/solvent system.

A variety of natural and synthetic pigments are used in paints, providing a complete spectrum of colors and a variety of finishes. Pigments are broadly classified as either organic or inorganic as shown in Tables [1](#page-3-0) and [2](#page-6-0), respectively. In organic pigments, the color is due to light energy absorbed by the delocalized p-electrons of a conjugated system.

The electrons do not usually absorb all wavelengths of light, so some frequencies are allowed to pass through. It is this unabsorbed light that we see, so the color of a given pigment is the opposite color of the frequencies absorbed by the molecule. The greater the number of conjugated double bonds in a system, the lower the energy of light absorbed; thus a molecule with little or no conjugation will absorb in the UV (and hence appear white), whereas one with more conjugation will absorb in the blue and appear yellow or in the green and appear red, etc.

Table 1 Some common classes of organic pigments

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Table 1 (continued)

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	Advantages	Disadvantages	Color(s)
Carbon black	High strength, good color, light and weather resistant	Thickens paint	Black
Titanium dioxide	High strength, high opacity, cheap, good UV resistance	Forms radicals that degrade the binder	White
Iron oxides	Light and weather resistant, unreactive	Cannot produce clean shades	Yellow, red, brown, black
Zinc chromates	Corrosion inhibition	Thickens paint	Yellow
Azurite (copper carbonate mineral)	Rich colors	Fades on contact with acid	Blue
Chromium oxides	Light, weather, alkali and acid resistant; thermally stable	Does not give clear colors	Green, blue
Cadmium sulfides	Heat and light resistant, clear pigment, high opacity	Expensive, poor weather resistance	Greenish yellow to red to bordeaux
Lithopone ZnS mixed with BaSO ₄	Pure tints when mixed with organics, unreactive, easily wetted	Poor weather resistance	White

Table 2 Some common inorganic pigments

Functional groups that absorb visible light (and hence appear colored) are called "chromophores," and some of the more common ones are.

 $-N = N -$, $-C = C -$, $-C = D$, $-C = S$, $-C = NH$, $-N = O$

Organic pigments are usually preferable as in general they are:

- Brighter
- Stronger (although this varies greatly between organic pigments)
- More transparent (this is not always an advantage)
- More stable

In addition, they have greater tinting strength (i.e., less pigment is required to get an equally strong color) and better gloss development, and some absorb UV light, preventing it from damaging the binder (Waldie [1974\)](#page-15-0).

However, inorganic pigments are also widely used as they do not bleed, are heat and light stable, and are much cheaper than organic pigments. In addition, they are used for some specialist pigments (such as anticorrosion pigments), and for black and white pigments, as it is not possible to get pure black or white organic pigments. One such white pigment, titanium dioxide, is widely agreed to be the single most important pigment in use today. It is the strongest known pigment in terms of both opacity and tinting power which coupled with its pure white tint and its fine particles size means that it can be used as an opacifier to prepare films with a high hiding power and reduced pigment content. This has resulted in paints with much improved elasticity and hence improved durability.

Considerations in Dye Design

Dye–Substrate Affinity

Dyes containing one or more azo groups (i.e., azo dyes) comprise by far the largest family of organic dyes. Prominent types are:

- 1. Acid dyes for polyamides and protein substrates such as nylon, wool, and silk
- 2. Disperse dyes for hydrophobic substrates such as polyesters
- 3. Direct and reactive dyes for cellulosic substrates such as *cotton, rayon, linen*, and paper.

Generally, the synthesis of azo dyes involves two steps. Step 1 is the conversion of an aromatic amine to a diazo compound (i.e., $Ar-NH_2 \rightarrow ArN_2^+$), a process known as *diazotization*, and **Step 2** is the reaction of the diazo compound with a phenol, naphthol, aromatic amine, or a compound that has an active methylene group, to produce the corresponding azo dye, a process known as **diazo coupling** (e.g., ArN_2^+ $+A$ r^{\prime}-OH \rightarrow Ar-N = N-Ar^{\prime}-OH). This process is suitable for forming both azo dyes and pigments. Typical structures of colorants that fall into the two groups are shown in Fig. [1.](#page-8-0)

Since the effectiveness of dyeing or printing process often hinges on the affinity between the dye and substrate, dyes are designed with a specific substrate in mind. In this regard, dyes must be designed that have:

- (a) Greater affinity for the substrate than the medium (usually water) from which it is applied
- (b) A high degree of permanence under end-use conditions (e.g., stability to fading upon exposures to water (wet fast) and/or sunlight (light fast))

The following is a summary of the types of considerations associated with the development of dyes for polymeric (especially fiber-based) substrates (Panda [2004\)](#page-15-0).

The availability of colorants for a specific substrate type is the result of a deliberate molecular design process that takes the target substrate and end-use application into consideration.

Dyes for Polyesters

Dyes developed for polyesters are known as *disperse dyes*. In this case, the mechanism of coloration involves "dissolving" the dye in the polymer matrix to form a solid–solid solution. Taking advantage of the well-known principle that "like dissolves like," disperse dyes are designed that are hydrophobic in nature. Such colorants are very sparingly soluble in water and derive their name from the fact that they are dispersed rather than fully dissolved in water to carry out the dyeing

Fig. 1 Some commercial azo dyes and pigments

process. An example is C.I. Disperse Blue 165 (Fig. 1). Disperse dyes have no affinity for hydrophilic polymers.

Dyes for Polyamides and Proteins

Dyes for polyamides and proteins normally form ionic bonds $-$ Fig. [2](#page-9-0) – within the polymer matrix. In this case dyes bearing a negative (anionic) charge are used because polyamides such as nylon and proteins such as wool, silk, and leather carry a positive (cationic) charge – especially during the dyeing process. Anionic dyes for polyamide and protein substrates are known as *acid dyes*.

An example of which is C.I. Acid Black 1 (Fig. [3\)](#page-9-0). They derive their name from the fact that they are typically applied to suitable substrates from a medium containing acid. These dyes have little to no affinity for polyester, cellulosic, or cationic polymers, since such substrates cannot form an ionic bond with them.

Dyes for Cationic Polymers

Dyes for cationic polymers also form ionic bonds within the polymer matrix. In this case, dyes bearing a positive (cationic) charge are used because polymers such as poly(acrylonitrile) carry a negative (anionic) charge in their backbone, making the ionic character of the interacting substances the reverse of that described above for acid dyes.

Cationic dyes for acrylic substrates were initially known as basic dyes, an example of which is C.I. Basic Red 18 (Fig. [3\)](#page-9-0). Today, they derive their name from the fact that they possess a cationic group. These dyes have no affinity for polyester, cellulosic, or polyamide polymers, since such substrates cannot form an ionic bond with them. However, cationic dyes can be used to dye protein fibers, and,

$$
\mathrm{D}\text{-}\mathrm{SO}_3^{\ominus}\mathrm{Na}^{\oplus} + \mathrm{Nyl}\text{-}\mathrm{NH}_3^{\oplus}\mathrm{Cl}^{\ominus} \xrightarrow{\hspace*{1.5cm}} \mathrm{D}\text{-}\mathrm{SO}_3^{\ominus} \text{ }^{\oplus}\mathrm{H}_3\mathrm{N}\text{-}\mathrm{Nyl}
$$

Fig. 2 Schematic representation of ionic bonding between dye (D) and nylon (Nyl)

Fig. 3 Structure of acidic black 1 and basic red 18 dyes

in fact, the first synthetic dye mauveine was a basic dye that was used for dyeing silk. This takes advantage of the presence of carboxylate $(-COO⁻)$ groups in silk and wool.

Dyes for Cellulosic Polymers

Cellulosic substrates include cotton, rayon, cellophane, linen, and paper, all of which are highly hydrophilic in nature. Therefore, require hydrophilic (water soluble) dyes for their coloration from a dye bath. In addition, dyes must be designed that maintain affinity when the substrate is exposed to water. This allows the color to remain on the substrate, when, for instance, a cotton fabric is laundered or a cup of coffee is inadvertently spilled on a sheet of paper containing printed information. The ease with which cellulosic substrates such as cotton swell and lose colorants during laundering has led to the design and development of more dye colorant families for cellulosic fibers than any other substrate. Dyes designed for cellulosic polymers are direct, azoic, vat, sulfur, and reactive dyes.

Direct dyes are so named because they were the first colorants that had affinity for cotton in the absence of a binding agent known as a *mordant*. Because these dyes are water soluble, many have low wet fastness. Figure [4](#page-10-0) illustrates two key properties of benzidine-based direct dyes, viz., (1) they tend to be linear molecules, and (2) they are able to gain close proximity to the cellulose chain, to maximize the effects of intermolecular interactions such as H-bonding. To enhance wet fastness on cellulosic fibers, methods were developed to apply water-insoluble dyes to cotton. Such dyes include those that are either water insoluble in their natural form or synthesized inside the polymer matrix as a water-insoluble dye.

The idea is that placing a water-insoluble dye within the polymer matrix would prevent color removal upon exposure of the substrate to water. In turn, this led to the development of sulfur and vat dyes (Figs. [5a](#page-11-0), [b\)](#page-11-0).

Fig. 4 H-bonding between cellulose (A) and a direct dye (B)

Vat dyes owe their name to the vatting process associated with their application, while sulfur dyes are so named because of the essential use of sulfur in their synthesis. The structures of vat dyes are rather well known, but sulfur dye structures are less well defined, because their bulky nature makes them unsuitable for standard methods for structure characterization. In their application, vat and sulfur dyes are converted to a water-soluble form that has affinity for cellulose and are subsequently converted back to their water-insoluble form, giving good permanence under wet conditions. Included in the family of vat dyes is the well-known natural dye *indigo*. While indigo is still the most important colorant for denim fabric, its typically small size causes this vat dye to be very susceptible to removal in a laundering process, giving jeans a faded appearance even after one wash. This underscores the importance of designing colorants for cellulosic substrates that possess the features required for them to remain within the polymer matrix when water swells the substrate.

A series of disperse dyestuffs derived from poly(amidoamine) dendrimer and thiazol derivatives were synthesized. The dyestuffs were finally applied for dyeing poly(ethylene terephthalate) fabrics in order to investigate their dyeing properties and the obtainable color gamut. The results indicated that the presence of poly (amidoamine) dendrimer as a core at the chemical structure of dyestuffs provided high build-up properties under high-temperature conditions. Most of the prepared dyestuffs displayed moderate light fastness and very good wash and dry heat fastness (Mohammad [2015](#page-15-0)). Anthraquinone-based solvent red, yellow (or green), and blue dyes used as essential ingredients were completely dissolved in styrene miniemulsion system, and a set of nano-colorants were successfully prepared by using a modified mini-emulsion polymerization process. In architecture, the obtained nano-colorants belong to a class of nanocomposite entities, in which a fraction of dye molecules are attached with cross-linked macromolecular chains and more dye being embedded in the interior of cross-linked polymer owed to the high hydrophobicity of dyes and phase separation between dye and polymer during the polymerization process. The performance characterization of the obtained nano-colorants indicated that they exhibited excellent chromatic properties attributed to the

Fig. 5 Chemical structures of (a) sulfur, (b) vat, (c) azoic, and (d) reactive dyes

nanoscale effects of homogeneous nano-colorants and really achieved superior migration fastness, light fastness, thermal stability, and good processibility (Zhenkun et al. [2008\)](#page-15-0). A series of novel naphthalimide dyes with amino or hydroxyl functional groups were synthesized by the condensation reaction of 4-bromonaphthalic anhydride with primary amines to form the naphthalimide, followed by regioselective aromatic nucleophilic substitution in basic media. In addition, fluorescence intensity decays of these dyes in dichloromethane were measured. The results suggest that various combinations of the dyes make good donor–acceptor pairs for energy transfer experiments. To anticipate the application of these dyes in the synthesis of dye-labeled polymers by radical polymerization, the synthesis of a monomer dye containing a methacrylate ester and a dye containing initiator for ATRP polymerization is described (Ghasem et al. [2008](#page-15-0)).

As stated earlier, pigments provide other properties to paints than just color and hiding power. Anticorrosive pigments are a very important area of study. Steel is probably the metal most often require protection against corrosion, and the development of pigments to inhibit the corrosion of steel has occupied chemists for many years. The anticorrosive pigment which has been used for many years is red lead, Pb3O4. In combination with linseed oil, red lead has in the past been the standard anticorrosive primer for iron and steel. Unfortunately, red lead is toxic, and much research had been, and is still being, carried out worldwide to find anticorrosive pigments which are as effective as red lead. Anticorrosive pigments for iron and steel commonly in use include zinc phosphate, zinc chromate, zinc molybdate, and barium metaborate, although alternatives to chromates are being sought as they are toxic and environmentally hazardous.

One special type of pigment has become of importance in recent years – the metallic pigment. Many modern cars have metallic finishes and this appearance is due to the inclusion of finely divided aluminum. Bronze may also be used as a decorative pigment. Zinc and lead powders may also be used as pigments, but in this case the metal powder has anticorrosive properties. Galvanic-type coatings containing zinc powder are an important tool in the protection of steelwork and are the subject of continuous research and development.

Requirements of Pigments for Plastics Application

The ability to produce the desired optical effect in the plastic product is obviously a prime requirement. However, the pigments must also be capable of withstanding the effects of the environment in which they are placed, both in processing and in their anticipated useful lifetimes.

A pigment will be selected for a particular application on the basis of its technical performance, but with due regard also to toxicological considerations and, inevitably, cost (Zollinger [2003\)](#page-15-0).

Optical Properties: Color and Opacity

The optical properties of materials are a result of combination of two effects arising from the way they interact with visible light: absorption and scattering. An object appears colored when it selectively absorbs certain wavelengths of visible light. The brightest, most intense colors are in general provided by the use of organic pigments. The colors of inorganic pigments are as a rule weaker and duller. High transparency in a plastic material requires the absence of light scattering centers either within the structure of the polymer itself or as a result of additives present.

To produce a colored transparent article, an inherently transparent polymer is colored either with dyes which dissolve in the polymer or with organic pigments. Organic pigments are generally low refractive index materials manufactured in a fine particle size form, much smaller than the wavelength of visible light, ensuring that they cause minimal light scattering and thus are highly transparent. When there is considerable light scattering, the plastic article will appear opaque. Inorganic pigments, a prime example being titanium dioxide, are usually high refractive index materials and therefore highly scattering, and so when incorporated into plastics, they provide opacity (Christie [1994\)](#page-15-0).

Fastness Properties

The heat stability of a pigment refers to its resistance toward changing color at high processing temperatures, and this is clearly an important factor for many plastics applications. Color changes resulting from inadequate heat stability, which can lead to off-shades or a failure to match shades, may be due to thermal decomposition of the pigment, to increasing solubility of the pigment at elevated temperatures, or to crystal phase changes. In the case of thermoplastics, heat stability is generally a critical feature in pigment selection (Hunger [2003](#page-15-0)).

The degree of heat stability required will depend not only on the processing temperature for the polymer in question, which can range from 150 to 350 $^{\circ}$ C, but also on the time of exposure. Heat stability requirements in the coloration of thermosets tend to be less severe than in case with thermoplastics. The properties of light fastness and weather fastness are clearly related although, on occasions, pigments which show good resistance to fading when exposed to light can perform less well under the combined attack of sunlight and moisture. These properties are determined principally by the chemical structure of the pigment, but they may also depend to an extent on its concentration in the polymer and on the nature of the polymer. Resistance toward changing color when exposed to acids, alkalis, or other chemicals is occasionally important for plastics applications (Christie [1998\)](#page-15-0).

Migration Resistance

Four aspects of migration of color from plastic materials may be identified, contact bleed, bloom, plate-out, and solvent bleed, all of which are associated to a certain extent with solubility of the pigment in the polymer. Contact bleed occurs when a colored plastic material causes staining of a dissimilarly colored material with which it is in contact. Pigments which show some solubility in the polymer or in a plasticizer present in the formulation are liable to give contact bleed problems as a result of molecular diffusion. Bloom is observed when a powdery deposit of pigment appears on the surface of a plastic product giving rise to poor rub fastness. This occurs when a pigment of limited solubility dissolves in the hot polymer, producing a supersaturated solution when cooled. In time, pigment molecules diffuse to the surface of the polymer and crystallization occurs. Plate-out refers to an accumulation of additive on the metal surfaces of plastics processing equipment. The reasons for plate-out are not fully established, but it appears to be a migration phenomenon associated with the presence in the polymer of a number of additives of different types, e.g., pigments, lubricants, and stabilizers. Solvent bleed occurs when color is leached from a plastic article when immersed in a solvent. The absence of solvent bleed is essential to ensure, for example, that a plastic bottle does not contaminate its contents (Zollinger [2003](#page-15-0)).

Electrical Properties

Pigments which reduce the insulating properties of poly(vinyl chloride) PVC are, for example, unsuitable for electrical cable, although such a defect is more often due to residual electrolyte on the surface of the pigment than to the pigment itself (Anon [1996\)](#page-14-0).

Dispersion Properties

Pigments are finely divided solid materials consisting of clusters of particles in varying degrees of aggregation. Two different types of particle cluster can be considered: aggregates and agglomerates. Aggregates are groups of primary particles (the smallest particles not normally further subdivided) joined at their faces. Agglomerates are considered to be groups of aggregates or primary particles joined at edges and corners. Agglomerates are much less tightly bound together than aggregates and are therefore easier to disrupt. During dispersion, agglomerates and to a certain extent aggregates are mechanically reduced to smaller aggregates and primary particles. It is essential that a pigment is well dispersed in a plastic material in order to achieve an even color distribution. For example, with organic pigments, color strength increases as particle size is reduced, so that efficient dispersion is essential. The term "dispersibility" refers to the ease with which the desired degree of dispersion is achieved (Christie [1998\)](#page-15-0).

Toxicological and Environmental Considerations

Plastics are generally perceived as relatively "safe" materials, so that it is essential to assess whether the incorporation of pigments introduces any hazards, either in the workplace or by exposure of the general public to the product. It is therefore reassuring that most pigments may be considered as relatively nontoxic, inert materials. It may be argued that the insolubility of pigments means that they pass through the digestive system without absorption into the bloodstream and so present little hazard. However, toxicological considerations are of critical importance for applications where ingestion is a possibility, such as plastics in contact with food and in toys and graphic instruments, and there is long-standing legislation limiting the use of certain pigments, notably those containing lead, hexavalent chromium, and cadmium, in such applications. The issues which are becoming of increasing concern to pigment manufacturers and users include environmental problems during manufacturing, such as the management of waste water and other residues, safety in production and hygienic material handling, and toxicological and ecotoxicological safety of the final products. The development of alternative pigments which offer equivalent technical performance, but which are more acceptable environmentally, is probably the most important challenge currently facing pigment manufacturers (Anon 1996).

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