Thinking about Ink: Composition, History, and Uses

Basic Components

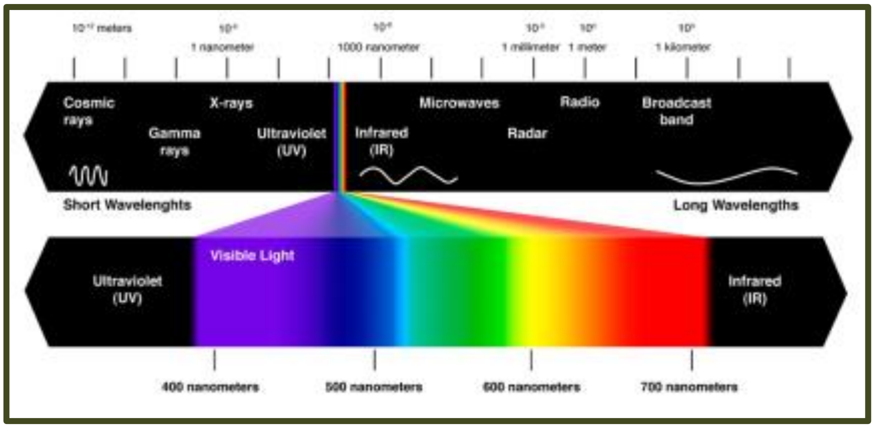
Ink is typically defined as a liquid of pigments and dyes used for writing and printing. Ink properties can vary greatly in terms of thickness, color, flow, and even permanence, however they are all generally composed of three main categories of chemicals: colorants, varnishes, and additives. Each component will be examined more in detail later, however, each name is indicative of their general purpose, where colorants provide color, varnishes are the medium through which the colorant is dispersed, and additives are additional chemicals added to vary ink properties such as thickness and flow. The different properties these components add can explain why your favorite pen writes so smoothly or why some black inks are blacker than others or why you avoid that one pen that always smudges. Additional work into this field has led to advancements such as quick dry pens and “erasable” inks, continuing to innovate with an ancient mixture of natural chemicals.

Colorants

The most directly observable component of ink is its color, and it’s what drives us to use a particular ink over another. While certain additives and types of varnishes are able to affect the physical appearance of some inks through changes in opacity, colorants are what make colors observable.

Wavelength and Color

First, it's important to understand how color can become visible on a chemical level. When we see color, we are just seeing the reflection of light take on a certain wavelength. Wavelengths correspond to the energy of the light with high energy light having shorter wavelengths. Thus when atoms absorb and release energy, it can come in the form of light with a discrete wavelength. The spectrum above shows the different types of light that can be emitted based on their wavelength. [2]. A small sliver of that spectrum falls into the visible light region, showing wavelengths that correspond to colors that we can perceive.



Light from an incoming beam is either absorbed or reflected. Reflected light is what we observe [1].

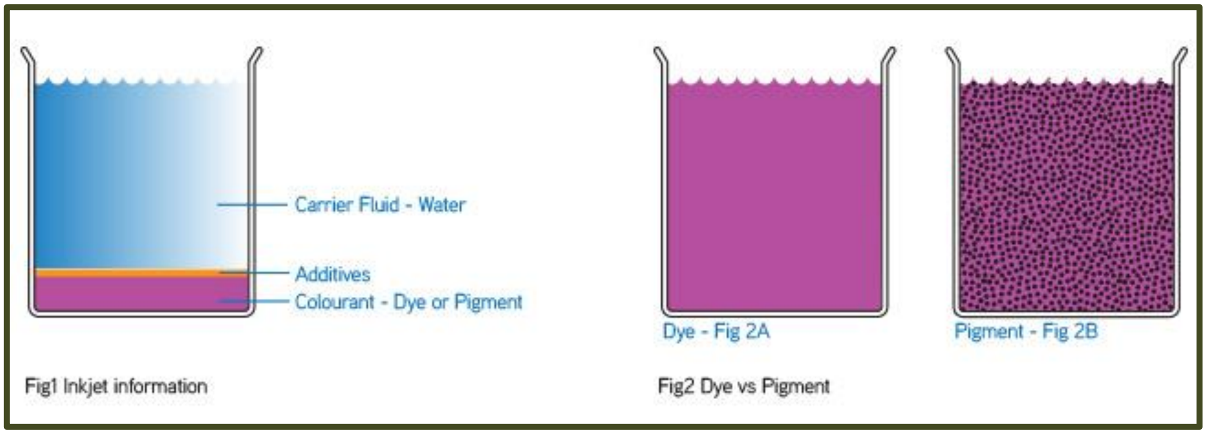
Materials do not possess color, rather, when light shines onto them, they absorb and reflect different wavelengths of light. The wavelengths that are reflected correspond to the colors that we see from that object. When we see a particular color of ink that is because the chemicals reflect a certain wavelength of light that corresponds to a wavelength and color within the visible spectrum.

Wavelength absorbed (nm)	Color absorbed	Color observed
400–435	Violet	Yellow-Green
435–480	Blue	Yellow
480–490	Green-Blue	Orange
490–500	Blue-Green	Red
500–560	Green	Purple
560–580	Yellow-Green	Violet
580–595	Yellow	Blue
595–605	Orange	Green-Blue
605–700	Red	Blue-Green

The above table shows the colors absorbed and observed at various wavelengths within the visible spectrum [4].

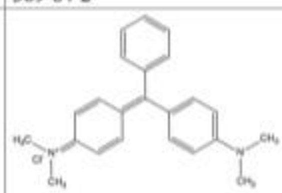
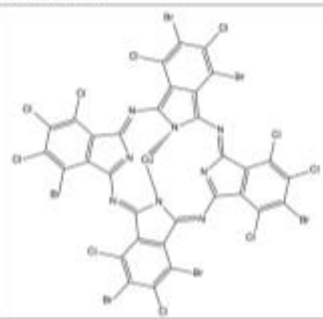
Dyes and Pigments

The chemicals that give ink these color reflecting properties can be classified as either dyes or pigments. The main distinction has to do with how soluble each chemical is. Generally, dyes are soluble whereas pigments disperse as particles within the varnish or medium they are placed in [4]. Pigments typically have longer chains and are more chemically complex making them difficult to dissolve. In addition, dyes are brighter than pigments, but they are less permanent [4].



The beaker on the left shows components of ink before mixing. The beakers on the right compare the dispersion of molecules in each solution once mixed [5].

The way in which they reflect light also varies as dyes reflect color based on chemical structure whereas a pigment's physical properties play a role as well. In fact, pigments reflect color through selective absorption and through scattering light [4]. Their solvent resistant nature also makes them more heat resistant, making them appropriate for more industrial use [4].

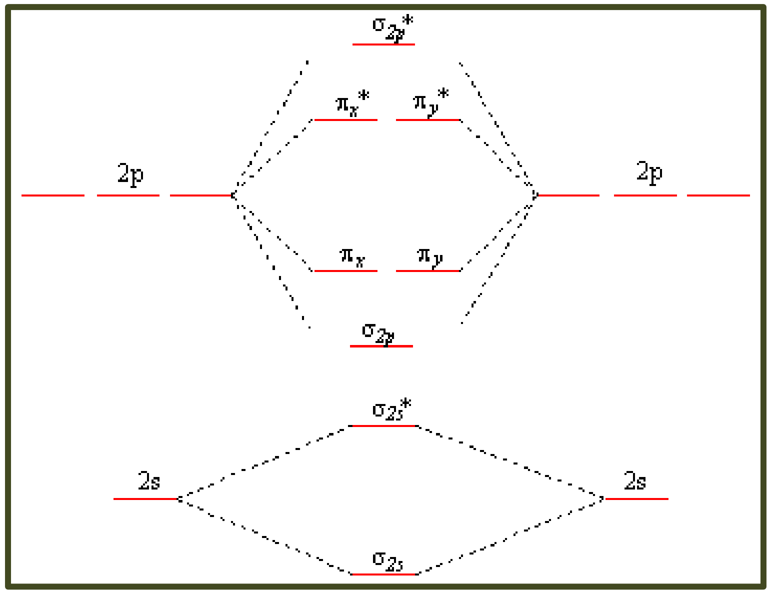
	Malachite green (basic green 4)	Pigment green 36 (phthalocyanine green YS)
CAS No	569-64-2	14302-13-7
Chemical structure		
Class	Triarylmethanes	Phthalocyanines
Chemical Formula	$C_{23}H_{25}N_2Cl$	$C_{32}Br_6Cl_{10}CuN_8$
Solubility	Very soluble in water	Insoluble in water
Physical form	Green crystals	Yellowish green powder

The table above compares the structure of a dye on the left with a pigment on the right [4].

While pigments can be dispersed in many types of varnishes, dyes only dissolve in solvents to which they have an affinity to. For example, polar dyes will only dissolve in polar substances and nonpolar dyes will only dissolve in nonpolar solvents.

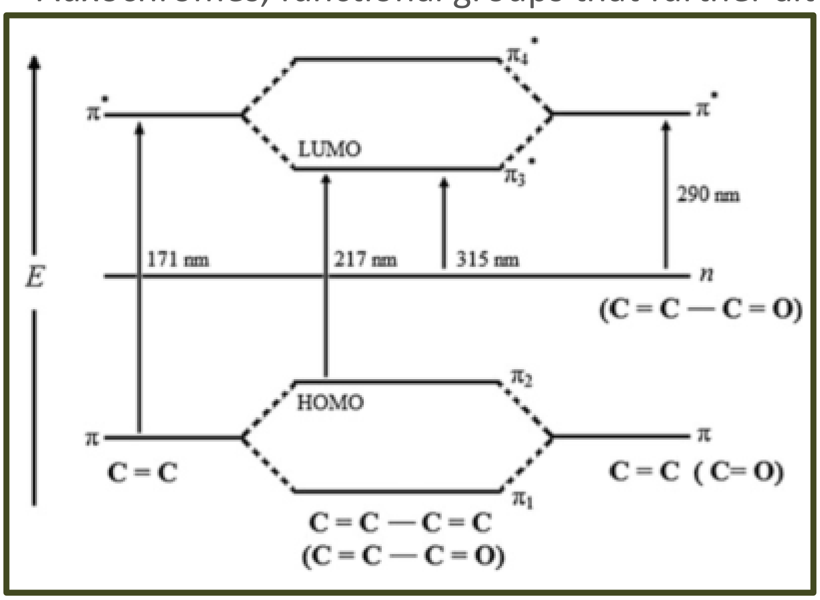
Chromophores

The color absorbing properties of dyes and pigments comes from chemical groups called chromophores. The colored properties of the chemical depend on where the chromophore is placed in the molecule. Chromophores will only cause color if it can absorb color with wavelengths of 400-800nm, falling in the visible region. Chromophoric groups include C=C, C=O, N=N, and NO_2. (See more complex chromophoric groups here.) Energy is absorbed and released from these groups when their electrons are "excited" to higher energy levels and then fall back to ground state. Energy levels become more complex when atoms are bound together. According to molecular orbital (MO) theory, these bound atoms can form σ bonding, π bonding, π antibonding, and σ antibonding orbitals that can experience electron transitions. Learn more about construction of MO diagrams here [8]. Essentially, electrons will jump from their highest occupied molecular orbital (HOMO) to their lowest unoccupied molecular orbital (LUMO). The energy difference between the HOMO and LUMO dictates what color is reflected.



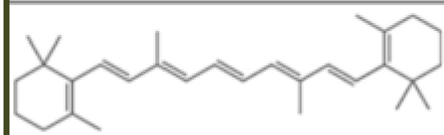
The diagram shows the arrangement of molecular orbitals as a result of bonding in order of energy, with the lower energy orbitals toward the bottom [6]. The asterisk (*) denotes an antibonding orbital.

Auxochromes, functional groups that further alter wavelength and intensity, can be attached to chromophores. The combination of an auxochrome with a chromophore produces a collective chromogen with different color properties. Auxochromes generally cause the HOMO and LUMO energy gap to decrease, resulting in an increased wavelength for the corresponding energy shift [4]. This occurs because the auxochromes



The molecular orbitals of each unconjugated system is represented on the left and right sides of the diagram, with the orbitals of the new, conjugated system in the middle. The wavelengths increase as the gaps in energy gaps decrease, resulting in a color change [4].

have non-binding electrons that can be shared with the original chromophore, extending the conjugation [4]. The extended conjugation results in new π_1 and π_2 bonding molecular orbitals and π_3 and π_4 antibonding orbitals that are held closer together than the pi bonds in the unconjugated system [4]. Because of this, the longer conjugated molecules have longer wavelengths with colors approaching the red end of the visible spectrum. The table below demonstrates this relationship, as carotenes with a greater number of conjugated double bonds produce colors with longer wavelengths [4] (see "Wavelength and Color").

Carotenes	Double Bond (C=C) Number	Color
 $C_{30}H_{44}$	7	Yellow
$C_{40}H_{56}$ (β-carotene)	11	Red-Orange
$C_{50}H_{68}$	15	Dark blue
$C_{60}H_{80}$	19	Black

More conjugation in carotenes results in longer wavelengths of light [4].

Additionally, the following image provides a visual representation of this phenomenon, as the more groups that are added to the system, the longer it gets, and the longer its wavelength becomes, showing how these groups can impact color [4].

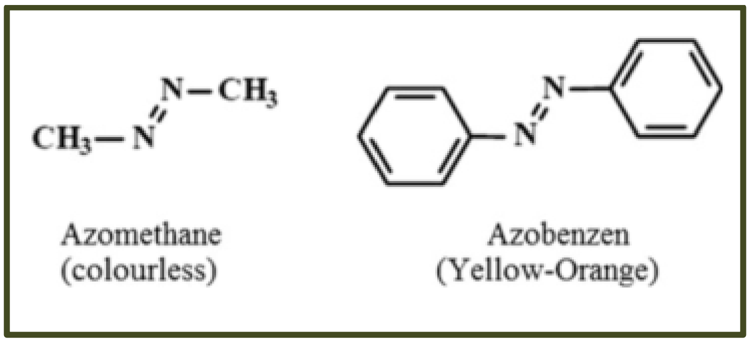
Conditions for Color

Due to these chemical concepts, organic colorants must possess certain components and properties to show color.

It must:

1. absorb light in the visible spectrum (see "Wavelength and Color"),
2. have a chromophore, and
3. be conjugated [4].

The importance of conjugated chromophores is seen in the image below where placing an azo group in between conjugated rings instead of between methyl groups produces a colored substance [4].



Varnishes

While colorants provide the visible color for the ink and are the most recognizable in the process, varnishes are the "backbone" of the ink produced because they are accountable for carrying the pigments and making sure that the ink has the indispensable properties that printers require. Varnishes have three main uses:

1. To be a solvent for the dye to dissolve in
2. To help the ink transfer from pen to paper
3. To bind the ink to the paper so it does not rub off

Varnish is typically a combination of a drying component, resin and a solvent. These different factors explain how the three main uses of a varnish are accomplished.

Drying Component

An ink in its supply form is a liquid but after application it is required to change to a solid. This change of state is referred to as ink drying and the varnish in the ink is responsible for this process. To think about this in laymen’s terms, the drying process describes how ink is transferred from pen to paper. An ink can dry in many ways but most commonly in one of three ways; Absorption drying, oxidation drying and evaporation drying.

In absorption drying, an ink dries when it penetrates by capillary action between the fibers of a substrate and when it is absorbed by the substrate itself. Capillary action is defined as the ascension of liquids through a slim tube due to adhesive and cohesive forces interaction between the liquid and the surface. Adhesive force discusses the forces of attraction between unlike molecules whereas cohesive forces describes the intermolecular bonding of a substance where its mutual attractiveness forces them to maintain a certain shape of a liquid. When intermolecular bonding of a liquid is inferior to a substances surface, capillary occurs. The ink remains a liquid but because of the degree of penetration it is effectively dry.

An ink dries by oxidation when the oxygen in the atmosphere chemically combines with the resin system converting it from liquid to solid. This process is very slow so often a fluid component is added that can be separated from the rest of the inks and is absorbed into the substrate which then leaves a film of ink on the surface which is dry to the touch but still soft and moveable. As autooxidation proceeds, the ink film becomes hard and tough. This process can be affected by the pigments and additives but is important to be controlled because it helps determine the rate of production and the quality of printing.

Some inks are formulated to dry by the physical removal of the unstable solvents from the ink formulation, leaving the resin behind to bind the pigment to the paper. The rate of evaporation depends on the affinity of the resin system for the solvents. Generally, the greater the affinity of the resin the slower the rate of solvent release. This affects the properties of ink in respect to the print-ability, the drying speed and the retention of solvents in the ink film.

Additionally, some inks used UV radiation as heating agent in the autoxidation process. There are other photochemical reactions which can be used to dry the ink, the majority dry using the ultraviolet mechanism. UV assists in the penetration of the varnish and is used to speed up the autoxidation of conventional inks.

Resin

Resins are complex organic compounds of high molecular weight. Resins should be film-forming, hard, colorless and soluble in the solvents to be used in the ink. The resin solids should increase as the viscosity of the varnish increases. On paper, resins can be chosen to give penetration into the substrate and therefor a low gloss level or less penetrating giving high gloss levels.

The gloss of an ink is its measure of its ability to reflect incident light and depends to a large extent on whether or not the ink forms a smooth film on the surface of the substrate and masks any irregularities. The degree of gloss depends on the nature of colorant, its particle size, shape and surface characteristics and the amount of resin and its ability to form a continuous film. In general, the more resin that is present in relation to the colorant, the higher the gloss will be. The resin determines how the gloss disperses the pigment and therefore is important because the better a pigment is dispersed the better the gloss is.

Different types of resins include acrylic resins which have low softening points, vinyl resins which give inks poor flow, gloss, and heat resistance, and hydrocarbon-soluble resins which yield high solid but low viscosity varnishes. A wide variety of chemical types of resin are used to modify the varnish in order to change desired properties.

Solvent

Solvents in ink are temporary ingredients that exist purely as a means of applying the varnish solids to the substrate by way of the printing unit. In theory, the solvent is then eliminated by evaporation or absorption and no longer is a part of the printing process. However, in practice this is often an over-simplification. The choice of solvents in ink is controlled by the resin system to be chose, the press speed, nature of the design, the substrate, end-use properties of the print and health and safety considerations.

The chemical nature of the resin system is the most important factor when choosing a solvent. The more resins blend with a solvent a lower viscosity will be given. Selection of solvents can be predicted by consideration of the

Solvent	Density at 4°C	Boiling point (°C)	Evap. rate (Bu Ac = 10)	Flash point (°C)	Hazard classi-fiction	General nature of risk	Risk phase Nos R	Safety phase Nos S
Acetone	0.79	56	115	−15(1°F)	—	Highly flammable	11	9 16 23 33
1,1,1,-trichloroethane	1.32	74	75	—	Class IIc	Harmful	20/22	2 25
Ethyl acetate	0.90	77	62	−5(24°F)	—	Highly flammable	11	16 23 29 33
Methyl ethyl ketone	0.80	80	57	−7(20°F)	—	Highly flammable	11	9 16 23 33
Hydrocarbon SBP 2	0.71	70−95	54	(−40°F)	Class IIa	Highly flammable, harmful	11 20/21 40	9 16 29 33
Isopropyl acetate	0.87	88	50	4(40°F)	—	Highly flammable	11	16 23 29 33
Hydrocarbon SBP5	0.73	90−105	35	(−7°F)	—	Highly flammable	11	9 16 29 33
Ethanol (74 op spirits)	0.79	75−81	33	13.9(55°F)	—	Highly flammable	11	7 16
n-propyl acetate	0.89	102	27.6	14(58°F)	—	Highly flammable	11	16 23 29 33
Isopropanol	0.80	81	23	12(53°F)	—	Highly flammable	11	7 16
Hydrocarbon SBP3	0.74	110−120	22	(15°F)	—	Highly flammable	11	9 16 29 33

Table of properties of possible solvents in ink.

solubility parameter and the possible hydrogen bonding associated with a solution of the resin in the system.

Solvent choice can also factor into the gloss of ink as described earlier because it affects pigment dispersion and the printing viscosity. If a solvent is not a true solvent, a poor flowing ink can result which can therefore affect the gloss.

History

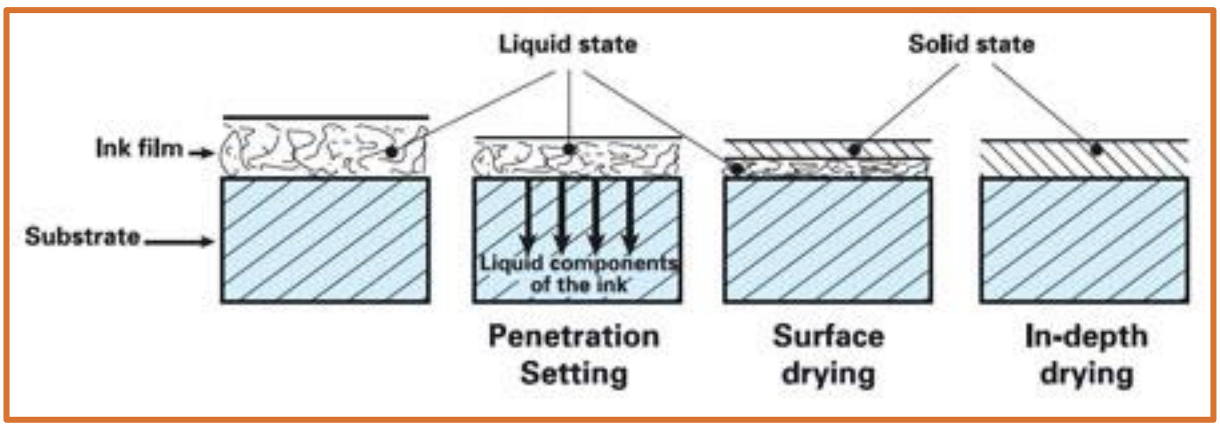
Using ink as a way to record information has been used for a very long time. In 300 AD, the Chinese developed a solid ink that was stored as a stick or cake-like material. To use this type of natural ink, one would have to shave off ink with a stick and then mix it with water. A 100 years later iron salts were used as a form of ink. This ink was special because it was mixed with tannin from gallnuts which would make the ink appear blue when first on appear but would then dry brown. During Medieval Europe times, branches would be cut and dried, then pounded and soaked in water to be boiled until thick and black. Then wine was added and dried in the sun in order to produce a thick ink. The issue with this ink was it would smear easily. Inks were then developed using soot in order to be used in the printing press, making an ink less susceptible to blurring. Today, new formulas for ink continue to be developed in order to create greater stability, better flow properties, and the use of natural materials at a lower cost.

Additives and Modifiers

Additives and modifiers are used in printing inks to make them more suitable for the job at hand. Wax can be added to increase the rub resistance of an ink used for packaging applications. Most inks for letterpress printing come ready-to-print without a need for additives to be added. Additives are often added to give the ink characteristics that varnishes and pigments cannot apply. For example, oils can give the ink certain flow and lubrication properties, waxes give the ink toughness and durability, and driers help promote rapid drying mechanisms. There are three major types of additives: drying agents, magnesium carbonate, and gel reducers.

Drying Agents

Drying agents usually react with the oxygen in the air to crosslink the resins used in the ink and they help to form a tough layer of ink on the surface of the substrate [17]. As soon as the ink is printed on the substrate, the ink begins to penetrate the substrate. Mineral and vegetable diluents



This diagram shows the drying agent process. As the diagram progresses through the steps, the ink is absorbed into the substrate [17].

contribute to how quickly the ink is absorbed. Typically, mineral diluents are absorbed quicker because they are more fluid than vegetable diluents. The type of substrate that is printed also affects how quickly the ink dries. The more porous a substance, the faster the absorbance. Substrates that cannot absorb the ink dry with air oxidation. Air oxidation is the use of oxygen to harden the varnishes in the ink. This process can be sped up with the use of additives. It typically does not take much of this additive to get the desired effect. Too much of this additive can inhibit the crosslink action. Crosslinking refers to the linking of different molecules by increasing the molecular weight of the ink film through a chemical reaction between the crosslinking additive and the resin binder [20]. The driers used most often are salts of cobalt and manganese.

Magnesium Carbonate

Magnesium carbonate (chalk) is another major type of additive and it changes the “length” of the ink [18]. It is used to modify the varnish to make it “shorter”. Magnesium carbonate is used to stiffen and shorten the ink. Ink tends to “string out” when the ink film is separated. Magnesium carbonate allows ink to be applied easily by the rollers to the surface of the image and when printed, it allows the ink film to break once the print is made and the plate begins to back away from the substrate. This gives a distinct edge to the image. Magnesium carbonate can add opacity to an ink. Typically, opaque white ink is used.

Gel Reducers

Gel reducers are the third major type of additive and it reduces the “tack” of an ink. Gel reducers are used when printing on soft paper stock which “picks”. When a substance “picks”, the paper fibers are pulled away as the plate or type pulls away from the paper stock [18]. The ink is tacky enough to separate fibers from the paper and these fibers can get mixed in with the ink or will tend to fill small open areas in the image. This requires frequent cleaning during a run.

Rheology Modifiers

Rheology modifiers affect a variety of ink properties. They can adjust the flow and leveling, help control the pigment settling, improve the penetration, and help control the temperature stability of the product. There are three major flow types: Newtonian, Pseudoplastic, and Dilatant [22]. Newtonian flow has changes in the shear rate but no change to the viscosity. Pseudoplastic flow occurs when the shear rate increases while the viscosity decreases. Dilatant flow occurs when the viscosity increases with the shear. Viscosity plays an important role in the behavior of an ink. The smoothness and the fluidity of an ink determines which printing method to use. Clays and organoclays are often used to alter the thickness of an ink [22].

Polymers

Polymers are another type of additive that contributes to the form of an ink. Polymers affect how an ink attaches to a substrate and they affect the ink coating film formed at a given temperature. They also enhance the print quality and abrasion resistance of inks. Polymers are classified by their conductivity, pH, viscosity, and molecular weight [19]. Molecular weight is one of the most important aspects of the polymers used in inks. A polymer with a high molecular weight has a high tensile strength but also has a high viscosity. The high viscosity is not ideal because the ink is not smooth and fluid. To counteract this effect, polymers with low molecular weights are used today. The film properties are not as sharp but the inks are more readily applied to the substrates. The melting point of a polymer determines the temperature at which the ink film forms. Polymers have a plasticity property to them that allows for them to be melted and shaped as desired.

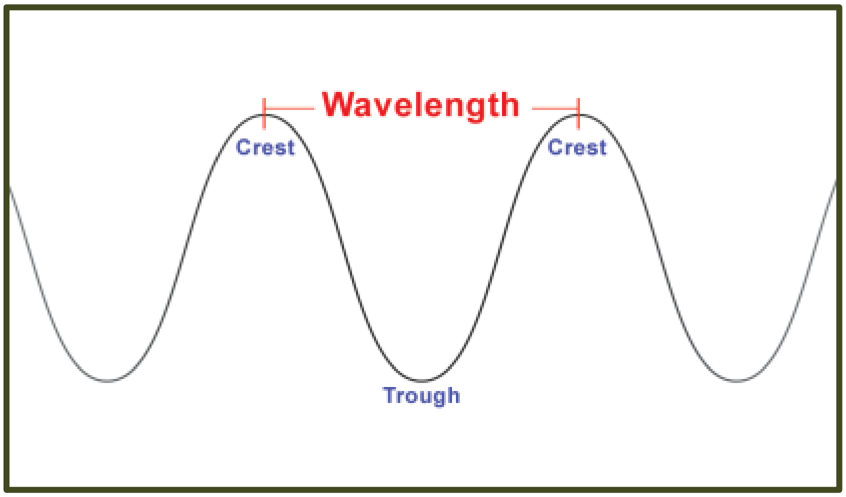
There are two different types of polymers: thermoplastic and thermosetting. Thermoplastics can be molded and remolded repeatedly and are easily separable and maintain mobility. Thermosetting polymers go through chemical reactions that result in infusible, insoluble bonds. These thermoplastics aid in the crosslinking properties of additives [19]. Polymers also determine whether or not an ink is a Newtonian or a non-Newtonian fluid as seen with the rheology aspect of inks. Polymers that affect the thickness of an ink typically have a high pH. These polymers have carboxylic acids groups and result from the ionization of acid groups. Not only do the inks themselves use polymers but the substrate printed on is often a polymer. Paper is the most common substrate associated with printing. Paper is essentially cellulose which is made from glucose: a polymer. Other substrates used include: polyethylene, polyamides, and other plastic materials.

Applications

Rheological modifiers are one of the most important additives in printing inks. The rheological modifiers adjust the ink composition to accommodate the different types of printing presses. Over 90% of inks are printing inks [23]. The color used in printing inks is imparted by pigments. There are many different types of print processes available such as: lithography (offset process), flexography, gravure printing, screen printing, letter press, and digital printing [23]. Each printing process requires a different type of ink varying in its viscosity and drying efficiency. Printing ink chemists try to make ink that does not clump by preparing a dispersion of pigment particles. Resins, preservatives, and wetting agents are also added to printing inks to acquire the desired consistency.

Glossary and Supplemental Content

Wavelength- the measurement of the length between a wave's crests to the next adjacent crest or from one trough to an adjacent trough. [3]



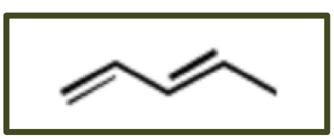
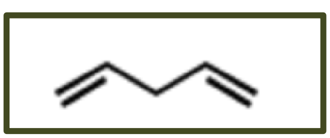
Inorganic compounds- Lack a central carbon atom and do not contain hydrocarbons (C-H)

Organic compounds- Based on a carbon chain covalently bonded to hydrogen, nitrogen, or oxygen

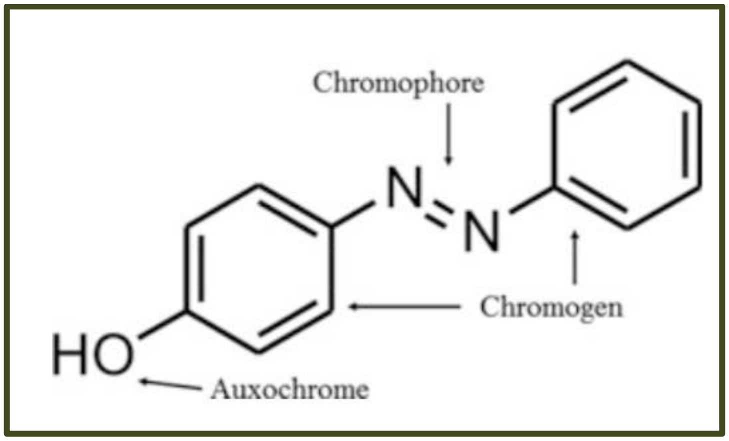
Energy Levels- Discrete values where electrons in an atom can exist

Ground State- The most stable electron configuration where electrons fill the lowest energy orbitals

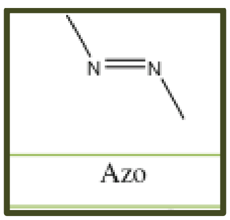
Conjugation- a molecule is conjugated if it possesses alternating double bonds and single bonds. Extended conjugation means that the chain of alternating single and double bonds becomes longer. The image on the left represents a conjugated system whereas the one on the right has adjacent single bonds and thus is not conjugated [7].

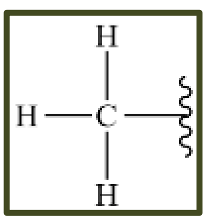
Chromogen- substance that can add color to a molecule when added to another compound. The combination of a chromophore and an auxochrome produces a chromogen as indicated in the image below [4].



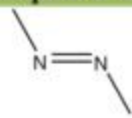
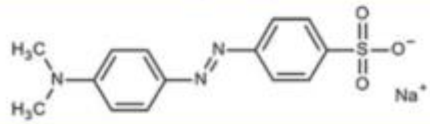
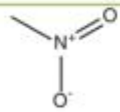
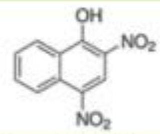
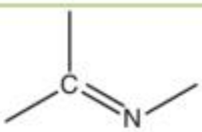
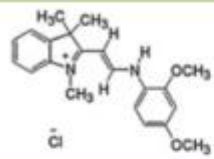
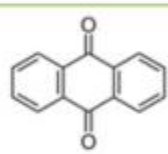
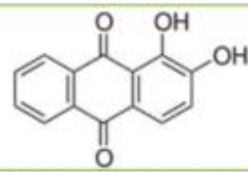
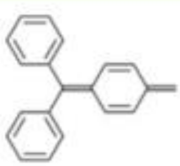
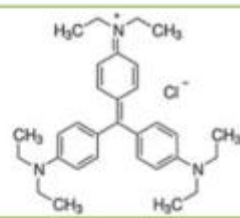
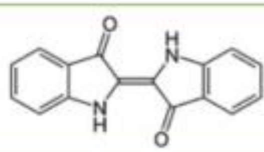
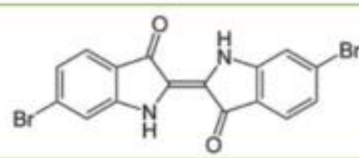
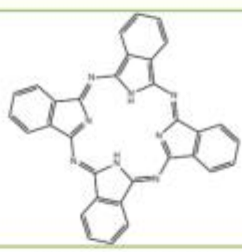
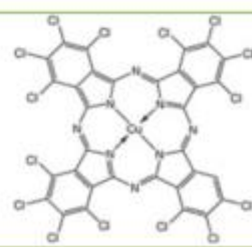
Azo group- functional group in which two nitrogens are double bonded to each other [4].



Methyl group- central carbon bound to 3 hydrogens [9].



More examples of chromophoric groups- [4].

Chromophoric Groups	Sample Dye
	
Azo	Methyl orange
	
Nitro	Martius Yellow
	
Methine	Basic Yellow 11
	
Anthraquinone	Alizarin (Turkey Red)
	
Triarylmethane	Ethyl Violet
	
Indigo	Tyrian purple
	
Phtalocyanine	Pigment green 7

Adhesive forces- The forces of attraction between unlike molecules

Autoxidation- The spontaneous oxidation of a compound in air (the presence of oxygen) and sometimes UV radiation to form peroxides and hydroperoxides

Capillary Action- Occurs when the adhesive forces are stronger than the cohesive forces which invariably becomes surface tension in the liquid

Cohesive forces- Intermolecular bonding where its mutual attractiveness forces them to maintain a certain shape of the liquid

Hydrogen Bonding- A relatively strong form of intermolecular attraction where a hydrogen atom is bonded to a strongly electronegative atom that exists near another electronegative atom with a lone pair of electrons

Solubility- The property of a solid, liquid or gaseous chemical substance (solute) ability to dissolve in a solvent

Substrate- A substance layer that underlies something or on which some process occurs

Viscosity - The measure of a fluid resistance to the ability to flow

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