Chapter Titles

• 1. Introduction: Colors Natural and Synthetic in the Ancient World
• 2. Discovery of the Physics of Color
• 3. The Chemical Causes of Color
• 4. Colorant Usage from Antiquity to the Perkin Era
• 5. Beyond Perkin
• 6. Major Analytical Techniques Based on Color: Volumetric Analysis; Chromatography; Spectroscopy; Color Measurement
• 7. Color on the Biological and Biochemical Front
• 8. Finale: Color in Foods, Photochemistry, Photoluminescence, Pharmaceuticals, Fireworks, Fun, and the Future
Chapter 1. Introduction

Colors, Natural and Synthetic, in the Ancient World

- Nature of Color
- Common Colorants in Antiquity
  - about three dozen in palette
- Blue – Egyptian Blue, Azurite, Ultramarine, Indigo
- Red – Vermilion, Carmine (carminic acid), Madder (alizarin)
- Yellow – Orpiment, Weld (a flavone from dyer’s rocket)
Grotte Chauvet
(southern France)

32,000 years before present
and 20,000 years older than the famous Lascaux cave
Oldest known chemical processes

- Manufacture of charcoal
- Roasting of hydrated iron oxide to produce various colors
- Purification of manganese dioxide
Carmine cochineal (carminic acid) – an historic pigment-dye from the New World

- The real treasure on the Spanish galleons
- Extracted from the egg sacs of female scale insects – cactus plant parasites (Dactylopius coccus)
- Successor to Old World cochineal from the Kermes insect
Rescinded April 19, 2012

New «natural» additive is lycopene, from tomatoes

Beetle coloring bugs vegans

Starbucks puts it in Frappuccinos

By Bruce Herbst
USA TODAY

Starbucks has been using a vegan coloring to what it recently began using to color its Strawberry Frappuccino beverage.

Marketing buzz — mostly due to its appeal in Mexico and South America.

Genes as that may sound, it’s a common, government-approved food coloring used widely throughout the food industry. It’s in everything from some brands of vanilla to those Kellogg’s Pop-Tarts flavors.

A vegan website, TheHillhicks.com, this month warned in a report that Strawberry Frappuccino is no longer vegan and how it is using the beetles for coloring. Starbucks made the switch in January when it aggressively moved away from artificial ingredients.

For Starbucks, which is eager to get artificial ingredients out of its food and drinks, it’s an unexpected PR problem. Frappuccinos in total, represent a $2 billion global business for Starbucks. Considering the beverage makes up a billion drinks a day in China, says Steve Siegel, founder, of The Hill Hicks, “It’s a huge issue for them.”

Phytochemicals like beta-carotene, which is used in some of the beetles, can also be found in tomatoes. That’s why this story is getting so much attention. Starbucks has been using the beetles to make its vegan Frappuccino.

New «natural» additive is lycopene, from tomatoes

Debella Fortney, co-founder of TheHillhicks.com, was informed of the change by an anonymous Starbucks employee.

She was at Starbucks to go back to using a vegan coloring like red beet, black carrot or purple sweet potato. She’s pointed a petition at her group on the website Change.org, under the heading "Starbucks: Stop using bugs to color your strawberry colored drinks," last Wednesday, it had 770 signatures.

"This was known as a drink that vegans can safely consume," she says, "we’re not trying to cause any problems. Our point is, vegans are detesting this and it’s not vegan."

But fortney says it’s simply moving to a different thing. "At Starbucks, we have the goal to minimize artificial ingredients in our products," says spokesperson Leslie Pasin.

Nutrition experts say it’s a good idea, but the results are mixed.

"Starbucks should be praised for getting out of artificial ingredients," says Michael Jacobson, executive director at the Center for Science in the Public Interest. But since some folks have plastic reactions to insects, he says, "raspberry flavored Starbucks should be colored with strawberries.

Markus Nieder, professor of nutrition at New York University, says she’s not concerned. "This is pretty far down on my list of outrageous food issues."
Vermilion (Cinnabar as mineral) $\text{HgS}$

One of the

- Most important
- Most ancient
- Most used
- And perhaps first synthesized pigments
Above: Ground cinnabar from a painter's pot found at Pompeii

«Tribute Horse & Groom» Chinese hand scroll, 1347
• (IV Dynasty – 4613-4494 BP)

• Calcium copper tetrasilicate, CaCuSi$_4$O$_{10}$

• Crystalline compound with glass impurities

• Exact method of manufacture still in doubt

**Egyptian Blue**
Excavations at Masada, Israel (2,000 years before present)

- Madder (from root of madder plant – alizarin)
- Yellow plant dyes from various sources
- Indigo (from plant Indigofera tinctoria) - $C_{16}H_{10}N_2O_2$
95% of the 55,000 pounds of indigo produced each year go into – guess what?
Tyrian, or Royal Purple
(chemically mostly 6,6’-dibromoindigo)

- Extracted from Murex snails
- 1 gram of dye = 10,000 snails
- So expensive that only royalty could wear garments dyed with it (anyone else risked a death sentence)
100 mg = 1,000 snails (natural product)
Synthetic product took less than one day to produce several grams (i.e., more than 30,000 snails!)
Ultramarine Blue
The Bamiyan Buddhas

Earliest known use of ultramarine
Fe(II) → Fe(III) + e-

«Prussian Blue» by Thomas Phillips, 1816

**Prussian Blue**
Chapter 2. Physics

Theories on the Nature of Light – Robert Hooke, Christiaan Huygens, Isaac Newton
Newton: Dispersion and Recombination - 1665
Early 19th Century

- Thomas Young – Interference
- Etienne Malus – Polarization
- Conclusion – Light has a wavelike character

1830 - 1865

- Michael Faraday – Charged particles produce magnetic fields; exert influence across space
- James Clerk Maxwell – Light is energy of a special form: A wave in a transverse electromagnetic field
  - \( \nu \lambda = c \)
  - \( E = h \nu \)

Theories leading to an understanding of the nature of light and the electromagnetic spectrum
1801-02: Johann Wilhelm Ritter discovered the ultraviolet

1800: Sir William Herschel discovered the infrared

Beyond the Visible
Chapter 3. Chemistry
Table 3.1. Timeline of discoveries leading to modern atomic structural theory

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<th>Discovery</th>
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<td>John Dalton (1766-1844)</td>
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<td>Dmitri Mendeleev (1834-1907)</td>
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<td>Cathode rays and their negative nature (electron)</td>
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<td>Sir William Crookes (1832-1919)</td>
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<td>Balmer series</td>
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<td>Electron (Crookes ‘cathode rays) charge/mass ratio</td>
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<td>J. J. Thomson (1856-1940)</td>
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<td>Alpha, beta, and gamma “rays”</td>
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<td>Isotopes</td>
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<td>Bohr model of atomic structure</td>
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<td>Niels Bohr (1885-1962)</td>
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<td>Neutron</td>
<td>1932</td>
<td>James Chadwick (1891-1974)</td>
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• The Chemical Bond

• Edward Frankland
• Walther Kossel
• G.N. Lewis
• Irving Langmuir
• Linus Pauling
• Electronic Transitions

• The Hydrogen Spectrum
• Balmer Series
• Color in Organic Compounds

• The MO model
• The VB Model
• The Free Electron Model
• O.N. Witt’s Empirical Observations
• Chromophore: any unsaturated grouping that is colorless
• Chromogen: an unsaturated system that is colored, or can be rendered colored, by attaching simple substituents
• Auxochrome: abandoned in favor of a less ambiguous term, “electron donor group”

Using these suggested terms, any atom that possesses lone pair electrons in conjugation with a \( \pi \)-electron system can be regarded as an auxochrome.

Using these revised definitions, one can conveniently classify the large diversity of colored organic molecules into four broad classes:
• \( n \rightarrow \pi^* \) chromogens
• Donor-acceptor chromogens
• Acyclic and cyclic polyene chromogens
• Cyanine-type chromogens
• Color in Inorganic Compounds

• Coordination Compounds – Pauling, Werner, Bethe, Van Vleck

• Chelates – Important in the dyeing process as mordants

• Charge transfer transitions

• Semiconductors
Chapter 4: Colorant Usage from Antiquity to the Perkin Era

- Body paint, face paint, hair coloring, cosmetics
This complexion powder contains about 13% bismuth oxychloride; MSDS is very non-committal; very little known about health effects.

This woman went blind; others actually died. The cosmetic: undisclosed aniline dyes

Pb in 400 brands of lipstick (C&EN 20 Feb 2012, p. 31)
• Ceramics, Glasses, Glazes, Stained Glass
• Pigment Use in Manuscripts
Pigment Use in Frescoes

The colors have been analyzed and contain the following: yellow is limonite, FeO(OH)\(n\)H\(2\)O; red is hematite, Fe\(2\)O\(3\), mixed with some galena, PbS; black is soot, C, mixed with iron compounds; green is a mixture of Egyptian blue, CaCuSi\(4\)O\(10\), and yellow ochre, Fe\(2\)O\(3\)H\(2\)O).
• Natural Dyes
Shellfish Purple: A Reprise
IS = Isatin; IND = Indigo; INR = Indirubin
Hexaplex trunculus Akhziv 290 nm

H. Trunculus Spain 300 nm

Bolinus brandaris Fiumicino 288 nm

Stramonita haemastoma Israel 288 nm
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<td>9.90</td>
<td>25.09</td>
<td>31,744</td>
<td>2.35</td>
<td>24.97</td>
<td>260,184</td>
<td>10.46</td>
<td>368,338</td>
<td>136,329</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>540</td>
<td>10,293</td>
<td>275,715</td>
<td>14.109</td>
<td>14.109</td>
<td>0.08</td>
<td>0.08</td>
<td>0.08</td>
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<td>0.08</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* Retention times based on the constant flow rate method, Method 1, of Table 1.
** Retention times based on the increasing flow rate method, Method 2, of Table 1.
<table>
<thead>
<tr>
<th>Murexide</th>
<th>Ammonium purpurate; 5,5’-nitrilodibarbituric acid monoammonium salt</th>
<th>1776</th>
<th>1818 (1853-1865)</th>
<th>CI 56085</th>
<th>Scheele (1742-1786) W Prout (1785-1850)</th>
<th>Uric acid treated with nitric acid and ammonia</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mauve</td>
<td>Mixture of four related aromatic compounds related to the safranines</td>
<td>1856</td>
<td>CI 50245</td>
<td></td>
<td>WH Perkin (1838-1907)</td>
<td>Impure aniline disulfate with potassium dichromate</td>
</tr>
<tr>
<td>Magenta (Fuchsine)</td>
<td>Rosaniline hydrochloride</td>
<td>1856</td>
<td></td>
<td></td>
<td>AW von Hofmann (1818-1892) J. Natanson, François Emmanuel Verguin</td>
<td>Impure aniline with stannic chloride</td>
</tr>
</tbody>
</table>
Route to Mauve
A.W. von Hofmann (1818-1892)
Adolph von Baeyer (1835-1917)

1876

1897
### Social Impact of the Dye Industry – A Juggernaut of the 19-20 Centuries

**Fashion.** Prior to aniline dyes, color austerity was the order of the day: only the rich could afford the rainbow of colors available in the natural dyes. But the ordinary person’s desire for the riot of color, now inexpensive and available to all, coupled with a clarity, variety, and fastness not found in the natural dyes, greatly stimulated dye research.

**The educational structure of industrial society.** The needs of the dye industry stimulated educational reforms that were already under way at the university level so that at the end of the 19th century, Germany had the finest system of scientific and engineering education worldwide – and this system influenced higher learning in all parts of the world.

**The social structure.** By employing large numbers of academically trained chemists in its plants and by growing a white-collar managerial proletariat, the dye industry displaced the independent tradesman and raised the educational level of the masses; this carried over into other disciplines as well.

**Political action.** The dye industry organized and supported lobbies, exerted strong influence on legislation, and helped establish a model patent system.

**Industrial research.** The dye industry originated and developed the industrial research laboratory and the research team, an organizational structure that prevails to this day.

**Power.** Through its hegemony in the dye industry, Germany became the foremost industrial power in Europe, leading to its almost becoming the foremost power in Europe. World War I was in actual fact a battle of technologies that promoted growth in other branches of the chemical industry, while the dye industry itself declined in importance.
• Color Naming and Classification
  
  • Names of Spectral Colors
  • Common Substances Named for Their Color
  • Elements Named for Colors (16)
  • The ‘Colour Index’
• Color Mixing and Color Specification Systems
  
• Hue – Color Circles
• Additive Color Mixing – Maxwell, the Impressionists
• Value and Chroma – Color Order Systems
• Major Analytical Techniques

Based on Color

Chapter 6

• Volumetric Analysis
• Chromatography
• Spectroscopy
• Color Measurement
Karl Friedrich Mohr
(1806-1879)

- ‘Father of Volumetric Analysis’
- Invented the ‘Liebig’ condenser, cork borer, Mohr burette, Mohr titration
- 1855 Textbook – plant pigments as indicators, concept of back titration, use of equivalents rather than moles
• differential adsorption on a stationary (solid) phase,
• differential solubility in a liquid phase involving more than one solvent
• displacement of one adsorbed species by another on a solid phase
• an adsorption series
• separation of components on a chromatography column
• elution of the various chromatographic zones
• qualitative analysis
• quantitative analysis

Mikhail Semyonovich Tsvet (1872-1919)

**Chromatography**
• The great chromatography success story was the development of liquid-liquid chromatography by Archer John Porter Martin (1910-2002) and Richard Laurence Millington Synge (1914-1994), who shared the 1952 Nobel Prize in Chemistry for this achievement.

1977 25-th Anniversary
Commemorative Stamp
Spectroscopy
Solar Spectrum:
- Mapped 570 lines
- Measured wavelengths to 7 sig.figs.
- Assigned major lines letters (e.g. D, which is the sodium-D line today)
- Used dark lines as reference points
- Future basis for Atomic Absorption Spectroscopy

Joseph von Fraunhofer
(1787-1826)
Color Measurement

Duboscq Colorimeter

Klett-Summerson Photoelectric Colorimeter
Vat Dyes – Reduction and Oxidation

Studies on the Color of Blood - “...the colouring matter of blood, like indigo, is capable of existing in two states of oxidation, distinguishable by a difference in colour and a fundamental difference in the action on the spectrum. It may be made to pass from the more to the less oxidized state by the action of suitable reducing agents, and recovers its oxygen by absorption from the air...” (George Stokes, 1864)
Colored molecules were his labeling, measuring, and diagnostic tools.

He used the products of the dye industry to argue for a new, chemical approach to biomedical research.

In his doctoral thesis of 1878 he outlined and classified the major synthetic dyes, resolved problems of commercial names and contaminants, and described how dyes could be used as staining agents to differentiate tissues.

He classified dyes into three groups: the primary amino dyes, the sulfonic acid derivatives of aniline blue, and the acidic dyes containing nitro and halogen substituents.

He learned how to exploit the loss of color as dyes were reduced to develop a tool to enable semi-quantitative measurements and provide information about cell surfaces – molecules of dyestuffs in his hands became probes and measuring devices.

Ehrlich learned to exploit the wide-ranging responses of vat dyes to reducing agents with two criteria: ease of reducibility and insolvability.
• Theory of Antibody Formation
• Chemoreceptors
• Chemotherapy
• Immunotherapy
• Monoclonal Antibodies

Ehrlich’s Monumental Legacy

Salvarsan (2005)
• Finale: Color in Foods, Photochemistry, Photoluminescence, Pharmaceuticals, Fireworks, Fun, and the Future

Chapter 8

• Foods
• Frederick Accum (1769-1838)
• Arthur Hill Hassall (1817-1894)

“In the preparation of sugar plums, comfits, and other kinds of confectionery, especially those sweetmeats of inferior quality, frequently exposed to sale in the open streets, for the allurement of children, the grossest abuses are committed... the red sugar drops are usually coloured with the inferior kind of vermilion. The pigment is generally adulterated with red lead. Other kinds of sweetmeats are sometimes rendered poisonous by being coloured with preparations of copper.” Accum’s Treatise of 1820
• Chief chemist at the U.S.D.A.
• Primary author of the 1906 Pure Food and Drugs Act
• Successor: 1938 Food, Drug and Cosmetic Act (FD&C)

Harvey Wiley (1844-1930)
Photochemistry & Photoluminescence
Nitrogen-rich, environmentally benign pyrotechnics are in the offing – the tetrazoles and tetrazines.

Boron is the answer to the BLUE!

Fireworks & Fun
- 15 December 1967: Silver Bridge Collapse
- Quantum Dots
- Multicolored Electrochromic Polymers
- Etc., etc.

The Future???????