# **A Method for the Identification and Assessment ofthe Presence of Bronzing in Printing Ink Films**

NPIRI Color Measurement Task Force"

# Keywords: colorant, colorimetric, ink, spectrocolorimeter, bronzing

Abstract: This paper examines the phenomenon known as "bronzing". The phenomenon has historically been ascribed to unusual morphological defects in the surface of an ink film. The most common explanation proposes that at some mass concentration, the individual pigment particles are pushed up through the surface of the varnish and light strikes the dry pigment resulting in a diffuse reflectance that differs from that observed when the pigment is surrounded by varnish. The measurement data reported in this paper clearly shows that this model cannot be correct. Not only do the extensive measurements reported here point to a more fundamental cause for bronzing, but they provide the framework from which to establish a measurement protocol for quantitatively assessing the level of bronzing in an ink film. This new framework can also provide the necessary additional information to a colorant formulation algorithm to allow the computer to predict at what concentration a colorant will begin to exhibit significant bronzing, either in a monochromatic ink or in a mixture of other colorants.

# Introduction

"Bronzing" is a phenomenon in which the appearance of a highly pigmented ink film changes dramatically as the direction of illumination or viewing is changed toward the specular or mirror direction. Various researchers have tried to define and specify the phenomenon. Portman (Portman 1931) seems to have recorded the first operational definition, "Some colors when converted into inks and

<sup>•</sup> R. Bassemir-Sun Chemical, G. Castillo-INX, M.Fuch-PMC, R. Incontro-NAPIM, K. Johansen-INX, T.Kummer-Superior Ink, C. Matarazzo-Pantone, D. Rich-Sun Chemical, M. Stanish-Penn Color, W. Zawacki-Flint Ink

printed have more bronze than others. This is a feature some what different than finish.". Foss (Foss 1938) expanded on the operational definition of Portman by stating, "In certain printing inks an unusually selective reflection may be noticed which varies considerably with viewing conditions. This is designated as bronze." Williams and Muller (Williams 1939) begin to qualify the general impressions stated by Portman and Foss stating, "Light reflected from the surface of the formulation with Prussian blue shows a reddish cast which is known as bronze." Finally, Buc, Kienle, Melsheimer and Stems (Buc 1947) provide the first comprehensive study of bronzing in printing inks, combining visual observation of the inks with spectrometric readings and photographic evidences trying to relate the phenomenon to film conformation. Following their descriptions, bronze is most easily observed if the ink film has been printed over a dark background, since this minimizes the reflection of the body color. It seems to have been called "bronzing" because the appearance frequently has a reddish brown hue and "sparkles" like a metallic surface. Bronzing is only observed in inks that are very highly pigmented. Figure l shows one possible illumination and viewing arrangement were a bronzing ink may be observed.



Figure l - One possible illumination and viewing geometry in which to observe or not observe the appearance of bronzing in an ink film.

Not every pigment will exhibit bronzing but those that are susceptible to bronzing will show the phenomenon both in single pigment inks and in mixtures of inks. Thus, a pigment may be observed to exhibit bronzing when used at 20% or greater absolute concentration in a vehicle but not at lower relative concentrations. If that pigment is then used in a mixture with a second pigment in the same vehicle, so that the total pigment concentration of the bronzing pigment is well under the 20% limit, one may still observe the first pigment to exhibit bronze appearance even though the total relative concentration of the bronzing pigment is below the critical concentration. Thus the development of bronze appearance is not based solely on the absolute amount of pigment in the ink.

Bronzing is both a problem and a desired objective. If a pigment, that is subject to bronzing , is used in the reproduction of an image it can produce unacceptable or unnatural appearance shifts. On the other hand, this striking, eye catching appearance shift may be just what a packaging designer needs to capture the attention and interest of a potential customer. It is desirable then to be able to identify the presence of bronzing and to predict when or how it will appear. Some common pigments that exhibit bronzing are: PB-61, PB-56, PB-27, PB-15, PR-49.2, PR-53.1 and PR-57.1.

# Theory

The most common theory as to the source of bronzing has been the hypothesis proposed by Buc, et. al. (Buc 1947), that if an ink is loaded with pigment particles and the morphology of the ink is such that a part of the pigment is pushed up through the surface of the vehicle the ink will have an "interface bronze" appearance. This is assumed to be because the diffuse reflectance of the pigment in air is different than the diffuse reflectance of that same pigment surrounded by the varnish. Figure 2 illustrates this possible explanation.



Figure 2 - Possible structure of a bronzing ink film.

Buc, et. al. asserted that this appearance is separate and distinct from the diffractive or interference effects seen in certain natural phenomenon such as oil films on water, insect casings, butterfly wings and bird feathers, even though the appearance is much the same. They termed this phenomenon interference bronze. Figures 3 and 4 are taken from the paper by Buc et. al. and show their measurements of a reflex blue and bronze orange ink using 0°:diffuse and 60°:60° specular geometry. Here curve C is the specular reflectance after overcoating the ink film with a layer of "grease". Note that the specular reflectance in their measurements has changed from the purplish bronze or yellowish red appearance of curve B, to a very pale blue or magenta appearance in curve C for both inks.



(specular excluded) reflectance factor of a reflex blue print; Curve B is the 60° specular reflectance factor of the same print; Curve C is the 60° specular reflectance factor of the same print over-coated with a layer of "grease" for a bronzing reflex blue ink.



Figure 4 - Curve A is the diffuse (specular excluded) reflectance factor of a reflex blue print; Curve B is the 60° specular reflectance factor of the same print; Curve C is the 60° specular reflectance factor of the same print over-coated with a layer of "grease" for a bronzing orange ink.

Elimination of the bronze appearance is not difficult in this model. If the pigment is protruding up through the surface of the varnish then overcoating the ink film with a layer of clear varnish should remove the bronze appearance. Indeed, that is the case. Appendix A shows the color readings of prints made from a Reflex Blue (C.I. Pigment Blue 61) pigment in a paste ink vehicle with and without a clear overcoating. When the relative concentrations are low (6.25%) there is little or no color difference but at high pigment concentrations (25% or 100%) bronzing is observed in the uncoated print and the color difference is substantial.

This present hypothesis would also seem to ignore the fact that when making the ink, the pigment is dispersed by wetting the pigment surface with a layer of adsorbed vehicle, thus displacing the air originally present, so that a pigment/air interface is no longer present, even if the pigment is so crowded as to break the surface of the ink film.

One of the more interesting properties of a bronzed coating is that the bronze appearance seems to vary as the specimen is rotated relative to the directions of illumination and view. At some angles the ink film has the bronze appearance while at others it has the same appearance as the non-bronzed film. If the pigment protruding above the varnish layer was in contact with air and was diffusely scattering the incident light, there would not be this strong angular dependence.

Suppose the pigment did not break the surface of the coating but instead was pushed up to the point where there is only a very thin film of varnish. As the film thickness reduces to less than 350nm the difference in refractive index between the pigment and the varnish forms a low resolution interference film, transmitting a range of wavelengths and reflecting a broader range of wavelengths. As the film is rotated the optical path length through the film increases and the film will either no long form a transmitted interference pattern or shift that transmitted pattern to longer wavelengths so that the reflected pattern will be in the near-infrared. Figure 5 shows an example of such a low resolution interference film. If this new hypothesis is correct then any instrumental reading which includes or is very near to the specular reflectance will contain some interference light and exhibit the bronze appearance. This experiment will attempt to test this new hypothesis.



Figure 5 - Proposed structure of a bronzing ink film.

### Experiment

To investigate ways to quantify the presence and source of bronzing a series of proof prints were prepared at the X-Rite Corporation, from a paste ink pigmented with reflex blue (Pigment Blue 61), a pigment known to have very strong tendency to exhibit bronzing at high pigment loads. According to the Pigment Handbook (Society of Dyers and Colourists 1982), PB 61 is an organic pigment with very little documentation on its physical and optical composition. It can be synthesized through several competing processes and has no consistent crystal structure. The paste ink used here, has an approximate pigment load greater than 20% weight of pigment to weight of binder. Inks films were prepared at the full pigment load ( 100% ), one fourth of the pigment load (25%) and at about one sixteenth of the pigment load (6.25%). The ink films were printed on both coated and uncoated stocks all with black and white contrast stripes. Table I shows the specimen coding and the properties of each of the fourteen prints used in this study. The substrates were various contrast test papers from the Leneta Company (Leneta 1987), 3NT-3 is a coated book stock, 3NT-4 is an uncoated stock, 3NT-7 is a newspaper stock and 105C is a lacquer coated card stock used for color matching. Each stock has a nominally white area and an area that was printed or dyed black. The bronze appearance is most readily seen when the diffuse reflectance from the substrate is reduced or absent. The ink films were over-printed with a various clear varnishes, some were low gloss, some medium gloss and some high gloss to see if the gloss level of the clear varnish would affect the varnishes ability to eliminate the bronze appearance.

After the initial specimens were prepared and characterized a second set of specimens were prepared. These specimens were offset inks prepared again with PB 61 beginning with the heaviest load possible in the ink vehicle, about 20% weight per weight of pigment to vehicle then decreasing the concentration down by steps of 10%. So, ten levels were prepared covering pigment levels of 100% of maximum to 10% of maximum loadings. A similar set was prepared using PR 53:1 similar to Pantone® Warm Red. Again, the specimens covered the range from Full Strength to 10% of Full Strength, where the full strength load was 27.25% pigment by weight. Copies of the data can be obtained from the authors.

The prints were characterized using several different optical instruments. Diffuse illumination and near-normal viewing was provided by a Datacolor International Spectraflash 600plus spectrocolorimeter (Spectraflash 600plus 1998), which is a traditional integrating sphere spectrocolorimeter with a white cap over the specular region of the sphere that can be opened and closed to either include or exclude the specular reflectance from a measurement, and by an X-Rite SP88 spectrocolorimeter (SP88, 1998). The SP88 is an integrating sphere version of the 938 which has a very high efficiency sphere surface and optimally includes the specular component.



Directional illumination and viewing readings were taken using an X-Rite 938 spectrodensitometer (938, 1998), which is a common 0°:45° geometry spectrocolorimeter designed to maximally exclude the specular reflectance, and finally using an X-Rite MA68 multiangle  $(45^\circ; -30^\circ, 45^\circ; -20^\circ, 45^\circ; 0^\circ)$ spectrocolorimeter (MA68, 1998). The MA68 is a multi-angle spectrocolorimeter designed for use in characterizing the color of automotive coatings that contain gonioappearance modifying additives such as metallic or pearlescent flakes. This instrument provides two viewing angles close to the specular direction and three away from the specular direction.

The two integrating sphere instruments provide the ability to take readings either with the specular reflectance included or excluded, while the spectrodensitometer collects readings that conform to the various ANSI CGATS.5 (ANSI CGATS.5, 1993) and ISO 13655 (ISO 13655, 1996) recommendations for densitometry and colorimetry of ink on paper. This series of instruments provides the ability to capture the spectral reflectance factors with and without the specular component at 8° and 45° off of the normal.

The spectral reflectance factors of the proofs were read with each instrument in the area of the white substrate, the black substrate, with and without the clear over printing. These spectral values were converted to colorimetric coordinates using the ASTM E 308 (ASTM E 308, 2000) tristimulus integration weight tables for CIE standard illuminant (ISO 10526, 1999) D65 and the CIE 1964

standard observer (ISO 10527, 1991). The tristimulus values were subsequently converted to CIELAB (CIE Publication 15.2, 1986) coordinates including metric chroma  $C^*$  and metric hue angle  $h^*$ . Where appropriate, color differences were computed for the CIE 1994 tolerance equation (CIE Publication 116, 1995) and for the CMC(l:l) tolerance equation (AATCC Test Method 173, 1992). The latter case was included due to its current popularity in setting many color tolerances but the l:c ratio was set at 1:1 since the specimens have very smooth surfaces with no apparent structure or texture. This reduction in the value of l:c ratio has been recommended by the CIE (CIE Publication 101, 1993).

#### Results

The tables of measurements in the Appendix illustrate that the proofs without overcoating, especially those printed on absorbing stock have significant differences in spectral reflectance factor curves and colorimetric coordinates as the load decreases from 100% to 6.25%. For the specimens with the overprinted varnish, the color differences are much smaller and rarely exhibit perceptible shifts in hue. The 0°:45° readings show little change for either coated or uncoated proofs. The same is true for the integrating sphere readings with the specular component excluded, though to a lesser extent. This is an indication that the bronzing is not a result of a diffuse absorption or an anomalous dispersion due to a resonance with the imaginary part of the complex refractive index. When one rotates the proof under a strong light source, a hue shift is perceived in the direction near to the specular direction.

If the data from the integrating sphere instruments with the specular component excluded or from the  $0^\circ$ :45° readings are subtracted from the specular included readings then a new spectral curve is obtained. If that new curve is then scaled to the same scale as the original readings and tristimulus integration applied, the resulting color coordinates, shown in Table A-2 in the Appendix, exhibit the same hue shift as seen in the visual observations. The same effect can be observed in the multiangle readings if the 45°:0° readings are subtracted from the 45° :-30° readings or if the integrating sphere readings of the over-coated proofs are subtracted from the uncoated proofs.

Figures 6 and 7 plot the spectral reflectance factor curves for specimen B of the first Reflex Blue set. Note that, unlike the report by Buc et. al., the specular reflectance factor curves of the varnished prints do not exhibit a pale blue color but are approximately spectrally neutral. In fact, the differences in reflectance factor between specular component included (SCI) and bidirectional (0°:45°) or between SCI and SCE are very similar to each other and very similar to the difference curves taken from the proofs that were varnished and those that were not varnished.

![](_page_8_Figure_0.jpeg)

Figure 6 - Spectral reflectance factor curves for specimen B of Reflex Blue ink measured on several different instruments and instrument geometries. All readings are taken over the black substrate but some of the proofs were overcoated with a clear varnish.

In Figure 8, we plot the differences between the spectral readings taken near to or including the specular component of the total reflectance (SCI or 45°:-30°) and those readings taken away from or excluding the specular component of the total reflectance (SCE,  $0^\circ:45^\circ, 45^\circ:20^\circ, 45^\circ:0^\circ$ ). Again, the curves are plotted for readings taken over the black substrate and for both the proofs with and without the clear varnish overcoats.

![](_page_9_Figure_0.jpeg)

Figure 7 - Spectral reflectance factor curves for specimen B of Reflex Blue ink measured on the X-Rite MA68 multi-angle, spectrogoniocolorimeter. All readings are taken over the black substrate but some of the proofs were overcoated with a clear varnish.

![](_page_9_Figure_2.jpeg)

Figure 8- Spectral reflectance factor difference curves for specimen B of Reflex Blue ink proofs. Differences are between those readings that include some or all of the specular reflectance and those that exclude most or all of the specular reflectance.

In Figure 9 are plotted the metric hue angle  $h^*$  versus the metric chroma  $C^*$  for readings taken over the black substrate of the full strength proofs, using readings from the multi-angle X-Rite MA68 instrument for the concentration ladder proofs of PB 61 and PR 53. Readings taken at the lower aspecular angles (15°, 25°) show a shift in hue angle as the concentration is increased, especially for the prints over the black area on the uncoated substrate. For the PB 61 series the hue shifts from the blue (330°) toward the red (370° or 10°) and for the PR 53 the hue shifts from the orange (70 $^{\circ}$ ) towards the yellow (100 $^{\circ}$ ). It is also interesting to note that the over black and over white prints for PB 61 for both coated and uncoated substrates converge to a single color at the lower concentrations while such a convergence is not observed in the PR 53 prints. This would imply that bronzing is present in the PR 53 prints at all concentration levels. Visually, the PR 53 specimens appear to show a yellow appearance when viewed against the black area of the uncoated substrates.

![](_page_10_Figure_1.jpeg)

Figure 9 - Polar plots of CIELAB metric hue angle  $(h^*)$  versus metric chroma  $C^*$ for the 100% load of Warm Red and Reflect Blue pigments, measured over the black substrate and for the varnished and not varnished sections of the proofs. The readings are taken from the *5* angles of the X-Rite MA68 with aspecular angles of 15°, 25°, 45°, 75°, 110° corresponding to measurement geometries of 45°:-30°, 45°:-20°, 45°:0°, 45°:30°, 45°:65°.

#### Discussion

The results above have shown several interesting features. First, the "bronze appearance" occurs only for observations (instrumental or visual) taken at angles approaching the specular direction. Absorption and dispersion can be observed in any direction, however there is a strong directional behavior in interference. More importantly, the interference is a surface, not a volume effect and the light reflected at or near the surface is only specular in nature and light of a different wavelength is transmitted into the film where it is available for absorption by the pigments or substrate. This is why the bronze appearance is most readily seen when printed over a black substrate. In that case there is no diffuse light being projected back up through the film and being additively mixed with the surface bronze reflectance.

Second, the "bronze appearance" always seems to appear in the long wavelength or red end of the visible spectrum. Figure 10, taken from the text Colour Science by Wyszecki and Stiles (Wyszecki, 1982) shows that as the angle of incidence is changed from directly over the normal to angles further from the normal, there is a shift in the in the center wavelength of the pass band of an interference filter and a broadening of the pass band.

![](_page_11_Figure_3.jpeg)

Figure 10- This figure is Figure 7(1.3.7) taken from the textbook *Color Science*  by Wyszecki and Stiles and shows the effect of changing the angle of incidence on the transmittance of a narrow bandpass, multi-layer interference filter.

In the present model there is not a uniform coating of varnish over the pigment particle, as in the case of an optically flat interference filter, but a range of thickness and shape as the film bends and conforms to the curvature or fracture of the pigment. In this case then each pigment particle has a range of wavelengths and a significant broadening of the transmitted band of wavelengths. The result is likely to be a low chroma, "yellow" or "orange" like behavior. This is similar to the way in which a near continuum of absorption bands results in a "yellow" appearance instead of a "green" appearance, peaked in the long wavelength end of the spectrum.

Third, reducing the pigment load or overcoating the ink film will usually eliminate the "bronze appearance". There have been reported cases where overcoating, even with several layers of clear varnish does not significantly reduce the bronze appearance. Under those conditions, the traditional model cannot begin to explain this departure from typical behavior. In the present model, however, we can speculate that either the pigment was assisted in the dispersion process by a surfactant that has a refractive index significantly different than either the pigment or the varnish and thus the 3 level refractive index difference is varnish, surfactant, pigment rather than the air, varnish pigment of the present examples. In this case, the dispersing aid must be changed to eliminate the bronze appearance.

Finally, if the readings taken at angles away from the specular direction or without the specular component of the reflectance factor are subtracted from those readings that include the specular component of the reflectance factor, the resulting spectral difference curve captures the perceived "bronze color".

Furthermore, the spectral curve of the "bronze color'' does not exhibit the sharp resonance structure of an anomalous dispersion absorption band. Rather, the color seems to move in from the long wavelengths and increases in intensity as the concentration of the pigment in the vehicle approaches the critical pigment volume concentration.

The CIELAB metric hue angle h\* illustrates the change in hue of the specular component difference curve. The CIELAB metric Chroma, C\* tracks the increase in pigment volume concentration. Thus, it should be possible to develop an index of bronzing that takes into account the increase in chroma and the shift in hue from the neutral level of a typical dielectric surface.

This effect of a color shift near the specular direction and no color shift away from the specular direction is exactly the same phenomenon that has been reported by researchers studying pearlescent (Gerlach, 1990) and interference (Wood, 1999) pigments used in modern automotive finishes and in security inks. Here the color shifts are more dramatic because the pigments themselves are the interference elements but in each case, the observed effect is a shift in hue from longer wavelengths to shorter wavelengths. In the case of bronzed inks, the shift also includes a shift in chroma because the non-bronzed state has the color of the light source which is translated to the neutral point by both the visual system and by the CIELAB color space metric. The bronze effect seems to disappear as the angle of view is rotated away from the specular direction since the optical path length through the thin film of varnish increases as a function of the cosine of the angular difference. Interference occurs when the film thickness is an integer multiple of half the wavelength of light. As the film thickness increases the wavelengths at which interference occurs shifts into near-infrared and outside of the spectral range of the human visual system.

# **Conclusions**

- 1. Bronzing is a phenomenon in which the appearance of a print of an ink made with certain pigments changes to an orange, metallic color when it is viewed at angles near the specular. Since this occurs only when pigment levels are high, current theory says that pigment particles are pushed above the film and exposed to air.
- 2. However, if the pigment did have an air interface, the strong angular changes in hue observed here should not occur. But the color of the bronze varies with the angle of viewing, even approaching the color of the non-<br>bronzed film. The distinction put forth in the literature differentiating The distinction put forth in the literature differentiating interference bronze from interface bronze is based on incomplete or inconsistent readings taken on a "home-built" spectrogoniophotometer and verified using models of dispersion and molecular spectroscopy that predate much of today's quantum chemistry.
- 3. Thick films of vehicle or overprint varnish will reduce/eliminate bronze. If the refractive index difference of the pigment and vehicle is small, bronzing disappears. As angles of viewing approach the specular, the bronze gets more intense and shifts in hue.
- 4. The currently accepted model of the cause of Bronzing is not supported by the measurements or the data obtained in this study. It is suggested that the presence of a layer of vehicle on the pigment particles of about 0.2 - 0.35 urn can cause broad-band interference colors in the light reflected at near the specular angle.
- 5.  $\Delta E^*$  of color measurements of a bronzed print and a section of the print overcoated with a clear varnish are proportional to the observed bronze. Difference curves between specular included and excluded are also indicative of the color, hue and intensity of the observed bronze.
- 6. Further work can now be done to better quantify the concentration at which bronzing begins. This will then allow computer-assisted formulation programs to highlight bronzing pigments and to either warn the operator to prevent bronzing or to optimize the formulation to enhance the bronze appearance while maintaining the desired body color.

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Table A-1 - CIELAB Coordinates for four of the Reflex Blue Specimens								
Sample	Location	OvrPrt	Geom.	$L^*$	$a^*$	b*	$C^*$	h*
B	Ovr / Blk	NoVar	<b>SCI</b>	37.78	5.85	$-41.41$	41.82	278.1
100%								
в	Ovr / Blk	NoVar	$0^\circ$ : 45 $^\circ$	27.85	7.27	$-52.74$	53.24	277.9
100%								
в 100%	Ovr / Blk NoVar		$45^{\circ}$ : -30°	35.49	7.44	$-5.98$	9.55	321.2
B	Ovr / Blk	NoVar	45°: -20°	14.38	3.35	$-6.92$	7.69	295.8
100%								
в	Ovr / Blk	NoVar	$45^\circ:0^\circ$	3.29	0.47	$-5.57$	5.59	274.8
100%								
B	Ovr / Blk	Var	SCI	25.77	$-0.03$	$-2.06$	2.06	269.0
100%								
B	Ovr / Blk $ $	Var	$0^{\circ}$ : 45°	2.63	$-0.03$	$-4.50$	4.50	269.7
100%								
B	Ovr / Blk	Var	$45^{\circ}$ : -30°	22.43	1.54	$-1.86$	2.42	309.5
100%								
B	Ovr / Blk	Var	$45^{\circ}$ : -20 $^{\circ}$	7.05	0.61	$-5.86$	5.89	275.9
100%								
B	Ovr / Blk	Var	$45^\circ$ : 0°	$\overline{2.24}$	$-0.09$	$-4.71$	4.71	268.9
100%								
E	Ovr / Blk NoVar		SCI	35.01	10.32	$-36.51$	37.94	285.8
100%								
E	Ovr / Blk NoVar		$0^\circ$ : 45 $^\circ$	29.95	4.90	$-38.61$	38.92	277.2
100%								
E	Ovr / Blk NoVar		$45^{\circ}$ : -30°	47.95	13.73	$-7.46$	15.63	331.5
100%								
E	Ovr / Blk NoVar		$45^{\circ}$ : -20 $^{\circ}$	32.86	9.72	$-7.69$	12.39	321.7
100%								
E	Ovr / Blk NoVar		$45^\circ:0^\circ$	18.26	4.84	$-9.90$	11.02	296.1
100%								
E	Ovr / Blk	Var	<b>SCI</b>	27.46	$-0.35$	$-5.32$	5.33	266.3
100%								
E	Ovr / Blk Var		$0^{\circ}$ : 45°	17.03	$-0.44$	$-8.01$	8.02	266.9
100%								
Е	Ovr / Blk	Var	$45^{\circ}$ : -30 $^{\circ}$	21.75	50.41	0.55	$-2.98$	3.0
100%								

APPENDIX A Measurement Results of Reflex Blue Ink Proofs

![](_page_17_Picture_160.jpeg)

![](_page_18_Picture_166.jpeg)

![](_page_18_Picture_167.jpeg)

 $\mathcal{A}$ 

![](_page_19_Picture_180.jpeg)