Accelerated Light-Fading of Some Historic Office Copy Documents

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ABSTRACT

The responsivity to light of the images and paper substrates comprising documents produced by a range of mostly twentieth century office copy processes has been investigated under accelerated exposure conditions using microfade testing. The results confirm that dye-based copies as a group tend to be fugitive, in some cases alarmingly so, but beyond this it is difficult to generalise. As might be expected, the responses of coated and chemically impregnated papers, many designed to be light and/or heat sensitive, is highly unpredictable and the microfade test results strongly reinforce the value of object-specific lightfastness evaluation rather than generalisations based on published data and assumed identity.

Keywords light, fading, display, office copying, documents, microfade testing, Thermo fax, facsimile

INTRODUCTION

For more than two centuries snowballing technological, industrial and social changes and the concomitant growth of commerce and public administration have fuelled the need to efficiently create and copy documents for communication, record keeping and archiving. Mundane as it may be, document copying is a cornerstone technology and this is reflected by the rapidity and ingenuity with which new technologies – optical, mechanical, chemical and electronic - have continually been adapted for the purpose (Batterham 2008). While most copies only see the light of day once or twice in their lifetime, a tiny tip of the vast iceberg of archived and accidentally preserved material acquires sufficient historic, social or personal significance to warrant display. Artists, including David Hockney and Warhol collaborator Gerard Malanga, have also created or reproduced images using mid-twentieth century copy processes like Thermofax and fax machines (Birmingham 2002, Norville-Day 1994). The relatively minor relevance of lightfastness to the documents' original purpose and the nature of the copying technologies have left us with records stretching back to the late nineteenth century that are potentially highly vulnerable to display lighting.

This paper describes observations from an opportunistic microfade testing survey of the lightfastness of a small number of documents held by the National Archives of Australia. It was thought to be interesting primarily because there is so little published information of its kind and because the documents are firmly attributed to particular copy technologies, if not necessarily representative of them or their class. A number of the processes are based on the heat or light sensitivity of chemically coated and impregnated papers, which years later still contain the same thermally and photochemically active components and/or their degradation products. The potential problems therefore not only include the light sensitivity of text and images but also the substrates that carry the images and provide contrast. These same chemicals may also be involved in photo-redox reactions that lead to cellulose oxidation, for example the catalytic role of iron compounds in photo-Fenton reactions (Thomas et al 2010).

In many of these cases loss of legibility and substrate deterioration is probably inevitable whether or not they are exposed to light, therefore the decision to display and if so for how long boils down to working out what is likely to destroy the documents first and how much additional harm a given amount of light exposure might cause. Unfortunately the relative rates of the different deterioration processes involved are essentially unknowable in practice without very long-term observation and measurement under ambient storage and display conditions. In this environment accelerated exposure is a useful tool, and perhaps the only one we have, but as this study clearly shows its results cannot be applied uncritically. Paul Whitmore, who invented the microfade technique wrote, "[t]he accurate prediction of the fading of different colorant systems is an elusive, perhaps unachievable goal" (Whitmore et al 2000), and for very complex systems in particular, like many of those described here, microfade testing's value is as an early warning system, identifying colourants at most risk of light-fading, and providing information about the probable nature of the colour change – whether the image or substrate is likely to become lighter or darker or more or less yellow with exposure. Combined with other information, accelerated exposure studies can also yield clues as to the photochemical mechanisms involved and assist with the evaluation of conservation treatments designed to mitigate light-fading.

METHOD

MICROFADE TESTING

Microfade testing is an accelerated light-fading technique in which the visible reflectance spectrum of a small area of a colourant (c.a. 0.3mm) illuminated by high intensity UV-free visible light is recorded as it changes. Colour change (ΔE_{00}), a perceptual measurement, is calculated from spectral change using standard equations, in this case CIEDE2000 (CIE 2001). The advantages of microfade testing are that the test area is small enough to fit comfortably within most typed and graphic lines, the method is rapid, and because it is essentially non-destructive real objects intended for display rather than surrogates may be tested. The Oriel Micofade Tester (Newport) employed in this study is substantially described by

Whitmore (1999) and the technique, including its application and limitations, has recently been reviewed in detail (Ford & Druzik 2012).

The illumination intensity of the UV and IR filtered light was approximately 6 million lux (MLux) providing 1 Mlux-hr cumulative exposure during a typical 10 min run. The temperature rise of the surface under test appears likely to be less than 5°C above ambient (Ford 2009), important in this case because some of the processes involve heat-sensitive materials. The quantitative validity of accelerated fading relies on the principle of reciprocity; that is that the extent of change depends only on the cumulative exposure, not the test illumination intensity, duration or whether it is intermittent or continuous. The fact that reciprocity is an unpredictable feature of colourant systems, rather than a characteristic of any particular accelerated test method, is one of the reasons for Whitmore's scepticism about the prospects for universal accuracy in fading rate predictions. Testing was sometimes repeated at 10% intensity (0.6MLux) to check whether reciprocity applied, at least over a single order of magnitude.

ISO Blue Wool Fading Standards (BWS) 1-3 were employed as internal standards and Blue Wool Equivalent (BWE) values in Table 1 were calculated from the colour change (ΔE_{00}) of samples compared to those of the BWs exposed under the same conditions. The BWSs range from the most light-sensitive at Blue Wool 1 (BW1) to the least at BW8, each successive step being approximately three times as lightfast as the one preceding it. Colourants less lightfast than BW3 are described in CIE157, *Control of Damage to Museum Objects by Optical Radiation* (CIE 2004) as having "high responsivity" to light for museum purposes. The cumulative exposure required to produce a change of one Just Noticeable Difference (Mlux-hrs/JND) in Table 1 has been calculated from the BWEs according to dose response data in the same document (CIE 2004, table 3.3) according to which, under UV-free illumination, 0.3 Mlux-hrs results in a JND (defined as $\Delta E00 = 1.6$), 1 Mlux-hrs for BW2, 3 Mlux hrs for BW3 and so on.

The experimental BWE values in Table 1 are the average of at least three replicates in each case, average relative standard deviation (RSD) 7.2% and the corresponding value for BWs 11.2%. Michalski (2013), who derived the dose response data in CIE157,assessed their uncertainty to be as large as plus or minus 1 BW step, or a factor of 3, therefore the uncertainty associated with the BWs' dose response is far larger than purely experimental errors involved in microfade testing.

DOCUMENTS AND COPY PROCESSES

Most of the documents in this study were collected by the National Archives of Australia over a number of years. The protocol developed by Batterham (2008, Appendix 1) was used to identify each copy technique. Most were dated, which assisted in their identification.

RESULTS AND DISCUSSION

In Table 1 the documents have been grouped into four categories on the basis of the type of colourant thought to be responsible for image formation. These are: dyes or pigments; ferric (pyro)gallate; metallic silver and Prussian blue (blueprints). Each covers a range of technologies that might otherwise appear to be unrelated; for example modern thermal printers, copy pencils, carbonless copy papers and spirit duplicates are all dye based; iron gall ink letterpress and the Thermofax front printing process both rely on ferric pyrogallate images; while the Thermofax back process is based on silver halide photography. Prussian blue (ferric ferrocyanide) stands apart because of its unusual response to light which, however, recent research has shown may be shared by ferric gallates in iron gall inks.

Within these categories however, the methods are differentiated by a variety of paper processing and coatings technologies and potential post-processing residues; for example different levels of unfixed silver halide, iron coordination complexes and salts in various oxidation states, unreacted dye precursors (for example azo dye components) humectants, stabilisers, clays and minerals, acids and alkalis and polymers plus any or all of their degradation and reaction products. There is ample scope for differences even between documents produced by a particular process because some of the earlier methods in particular involved sequences of reagents (fresh and potentially exhausted), washing and fixing steps, different suppliers of dyes and papers and so on. Finally tint strength (and prior fading) can also lead to variations in the fading rate of a given colorant of as much as one to two blue wool steps (Michalski 1997).

It is important to note the nature of the colour change in Table 1 as well as its extent because not all colour change is "fading". Images that darken and papers that become lighter and/or less brown (chroma loss) do not constitute a legibility problem, although their responsivity may be a sign of unreacted light-sensitive precursors. Almost all uncoated papers exposed to UV-free light "bleach" (less yellow and either lighter or darker) and although this process may temporarily improve contrast, yellowing will subsequently resume in the dark, perhaps more rapidly because of the prior light exposure (Forrskahl & Maunier 1993). Coated and chemically treated papers, especially where they are designed to be heat or light sensitive, are unknown territory but it is clear from even these few examples that in some cases gross colour changes leading to loss of contrast have taken place in the dark.

DYE BASED COPIES

In general terms dye based processes – which include thermal copies of various kinds, spirit duplicates, diazotypes, carbonless copies, copy pencils and others not represented here like "aniline" dye letterpress, hectography, stencil processes, non-carbon typewriter inks and "carbon" papers are potentially very fugitive. In the absence of reliable document-specific fading rate information – meaning microfade testing or colour monitoring whilst on display (very difficult) - their lightfastness as a group should be assumed to be at least equivalent to BW2, however it is difficult to generalise even on the basis of the method. The two direct thermal print copies, a 1998 thermal fax and a supermarket receipt from 2012, for example, are very different (Figure 1). Mostly, like the blue wools, colourants fade more slowly as fading progresses and they often plateau when light-sensitive components have been destroyed early on. However, in the example shown in Figure 7, the fading actually speeds up as it progresses, and as the

Process		Text/Image			Paper		
	Image type	Colour	BWE	Colour change	Colour	BWE	Description of colour change.
Copy pencil (CP1)	“aniline” dye	purple	<<BW1	lighter, chroma loss	n.a.	n.a.	n.a.
Spirit duplicate 1950’s	“aniline” dye	purple	<<BW1	lighter, chroma loss	n.a.	n.a.	n.a.
Carbonless copy 1972	leuco dye	blue	2.8	lighter, less blue	yellow(ed)	1.5	lighter, chroma loss (less yellow).
Carbonless copy 1985	leuco dye	purple	<BW2	lighter, chroma loss	green	2.8	lighter, chroma loss, complex
Thermal fax, 1998	Leuco dye	grey	<<BW1	lighter, chroma increase (browner)		2.9	lighter, chroma loss (less yellow)
Thermal printer, 2012	?	black	2.3	lighter, browner	white	2.9	lighter, chroma loss (less yellow)
Diazotype 1976	azo dye	black	1.8	lighter, chroma increase (yellowed)	brownish	2.3	lighter, chroma loss (less yellow)
Electrofax, 1972	dye or carbon	grey	>3.0	no change	white	2.4	lighter, chroma loss (less yellow)
Ferrogallate architectural print, 1911	Fe(III)-gallate?	black	2.2	darker, chroma loss (less yellowed)	brown	1.6	lighter, chroma loss (less brown)
IR sublimation, 1960’s	Fe(III)-gallate	brown	1.8	darker, rapid initial response, then very little change.	brown	2.2	darker
Thermofax FP, front process, 1961	Fe(III)-gallate	black	1.9	lighter, chroma loss.	very brown	1.7	lighter, chroma loss (less brown)
Thermofax BP, back process, 1965	silver	black	1.6	lighter, complex chroma and hue changes	brown		lighter, complex chroma and hue changes.
Dual spectrum, 1967, yellowed paper,	silver	grey	2.7	lighter	yellowed non yellowed	1.6 2.7	Complex. Yellowed paper bleaches, non-yellowed may yellow with exposure.
Kodak thermal, 1961	silver	brown	>BW3	Darker, less yellow(ed)	brown	2.9	darkens slightly
Diffusion transfer, n.d.	silver	brown	2.4	darker, chroma loss (less yellow(ed).	brown	2.8	darkens slightly, complex,.
Blueprint n.d..	Prussian blue	blue	-	lighter, chroma loss but colour reverts overnight.	-	-	

Table 1. Summary of microfading results grouped by image type. The Blue Wool Equivalent (BWE) is the rate at which the colour change occurred compared to the ISO Blue Wool Fading Standards exposed under the same conditions.

insert shows beyond 1Mlux hr it proves to be much more fugitive than BW1. Increasing rate (Type V) fading curves of this kind have been explained as large dye aggregates breaking down into more vulnerable smaller particles, probably as a result of heat of illumination (Cox Crews 1987). This explanation is unlikely in this or the other example of the phenomenon in Figure 5, not only because temperature rise is probably less than 5°C, but also because in the latter case exactly the same pattern and degree of colour change/Mlux-hr was observed when the illumination (including residual IR) intensity was cut to 10%.

Where documents contain the ubiquitous purple inks used for official stamps, purple “carbon” copies, typewriter ribbons, copy pencils, hectographs and spirit duplicates from the late nineteenth century onwards they should be assumed to be worse, perhaps much worse, than BW1. They can be seriously

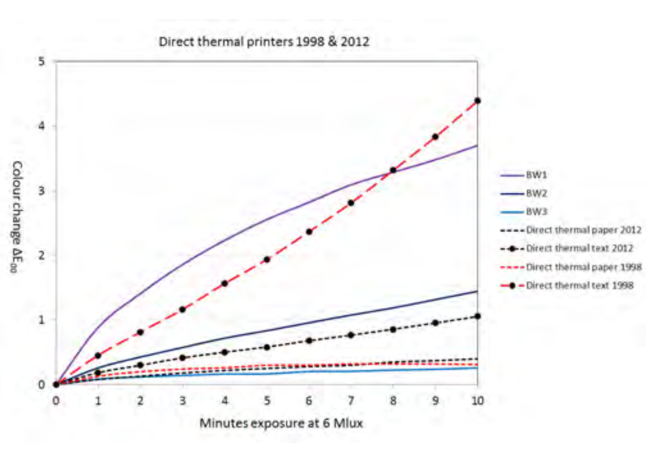

Figure 1. Colour change curves of the image and paper areas of two different modern direct thermal printouts during a 1Mlx-hr exposure.

faded in a few months, to a years display, even at 50 lux with UV rigorously excluded. Figure 2 shows the microfading curves of 11 examples of "purple inks" or "purple official stamps" from documents from the late nineteenth to the mid-twentieth century[1], including the spirit duplicate (1961) in Table 1. These inks, usually described as "aniline" dyes (e.g. Batterham 2008, p.49), probably contain one, or a mixture, of the structurally related compounds variously known as Crystal Violet, Gentian Violet, Victoria Blue and by many other names. The same compounds are also found in modern ball point pens [2] (Crown 1961). They rapidly achieved popularity as the basis for dye-transfer copy inks because of their extraordinarily high tinctorial power which allowed many copies to be run off each master. Figure 3 provides the same data for the ink from three twentieth century copy pencils, two purple and one cyan (the problem is not confined to purple), including the example from this study (CP1). A reciprocity test showed that CP1 faded twice as much for the same cumulative dose (1Mlux-hr) at a test intensity of 0.6MLux as it did at 6Mlux, suggesting that it is even less lightfast in practice than the accelerated testing indicates.

The "carbonless" copy examples (Figure 4) are interesting for different reasons. The first of these documents (1972) is instructive because, although the coloured leuco dye (activated by an acid layer on the copy sheet) is comparatively good at about BW3, a green printer's ink on the pre-printed form fades at a rate equivalent to approximately BW1. This is relatively unusual for modern printers' inks and reinforces the point that generalisations and assumptions about fading rates are often wrong in practice and that microfade testing, particularly of high significance documents, provides much more certainty (Ford & Smith 2009). The second (1985) carbonless copy on a green, relatively lightfast, paper is notable because of the unusual, but reproducible, fading progression of the dye (Figure 5). Multi-step fading curves may be a sign of mixtures of colourants with different fading rates or the formation of coloured intermediates during the photochemical reaction. The accelerating rate means that like the thermal fax in Figure 1 it would eventually outpace BW2.

BLUEPRINTS (PRUSSIAN BLUE)

Blueprinting is a photographic technique which uses the photoreduction of iron(III) to iron(II) to produce either a positive or negative image composed of the deep blue complex ferric ferrocyanide (Ware 2003). A fading rate has not been given for the blueprint in Table 1 because although

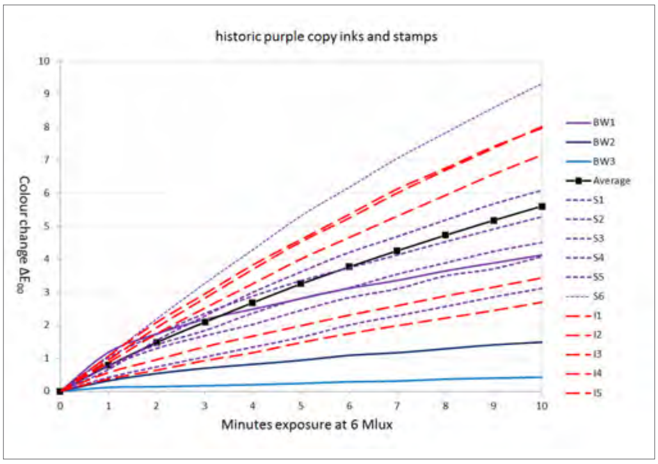

Figure 2. Colour change of 11 purple inks used on official stamps (S) and copies (I) from the late 19th to mid-20th Centuries.

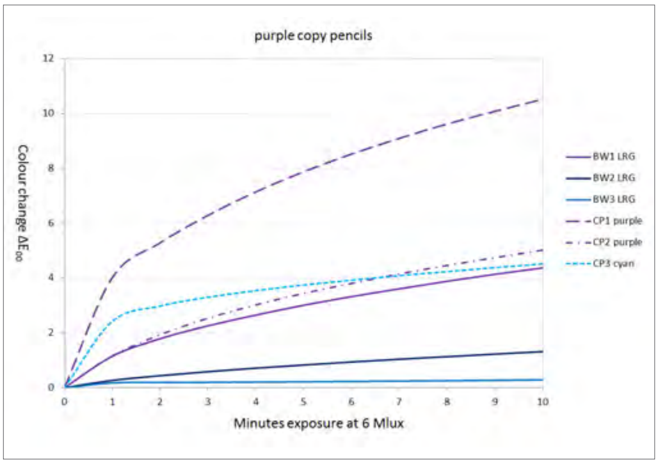

Figure 3. Colour change of purple and cyan dyes from three 20th C copy pencils.

Figure 4. "Carbonless" copies, 1972 (left), 1985 (right). The green printers' ink on the left hand copy is far more fugitive than the blue copy ink.

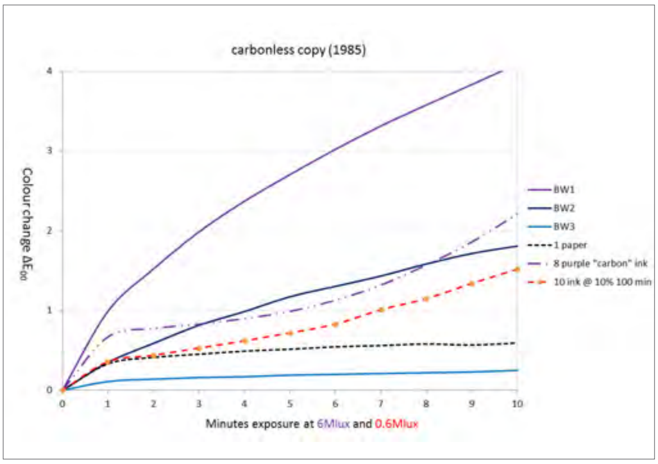

Figure 5. Colour change curves of the 1985 carbonless copy in Figure 4, with the result of a test at 10% illumination intensity over 100 minutes, demonstrating reasonable reciprocity between 5Mlx and 0.5Mlx

it responds extremely rapidly to continuous illumination under accelerated conditions, the fading spontaneously reverts post-exposure if oxygen is available. This is illustrated in Figure 6. Photoreduction of Fe(III) to Fe(II) within the complex in the presence of oxidisable impurities results in a colourless compound (Berlin white) with a similar structure, which molecular oxygen dynamically re-oxidises to Prussian blue. Under accelerated conditions photoreduction outpaces oxidation, but at ambient light levels re-oxidation probably keeps up with the fading. Although in Figure 6 overnight reversion is incomplete, studies carried out by Ware (2003) indicate that fading does not build up with successive cycles under more normal lighting conditions, presumably meaning blueprints and cyanotypes are essentially lightfast. This is an extreme example of reciprocity "failure", which in this case vastly over-estimates the fading rate of the colourant.

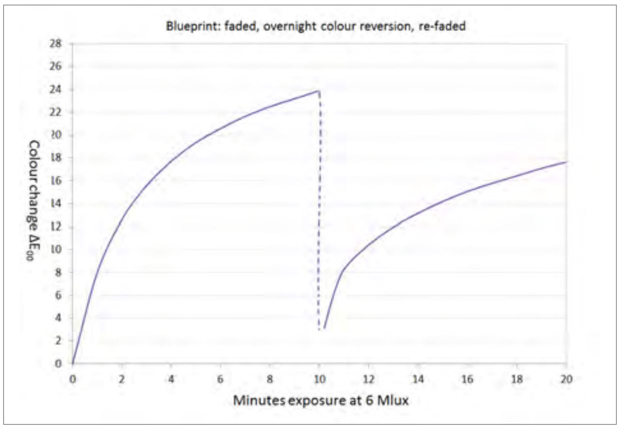

Figure 6. Experiment demonstrating overnight colour reversion of a blueprint. The dashed line represents 15 hours darkness during which time most of the colour lost to the initial accelerated fading has returned.

FERRIC (PYRO)GALLATE COPIES

The coordination compound(s) responsible for the colour of Thermofax FP (front process, 1961), ferrogallate (1911) and IR sublimation (1960s) prints are assumed to be similar if not identical to the ferric gallotannates responsible for the colour of iron gall inks and letterpress copies from iron gall ink originals (e.g. Jackson 2012). This is why they have been grouped together in this paper, however the results show that this assumption may not be relevant. Most iron gall inks become lighter and more yellow/brown during uninterrupted accelerated exposure, a pattern which the IR sublimation (1960s) copy, for example, does not follow. In this case both the image and the already brown paper become browner, but they both darken rather than become lighter. Initially the image darkens much more rapidly than the paper, which in practice (assuming reciprocity) would probably help to maintain legibility by maintaining contrast (Figure 7). Further down the track, however, after the response of the image essentially plateaus, the paper continues to darken and brown at a steady rate, reducing contrast if left on display long enough. The colour changes undergone by the (brown) paper and darker image areas of the Thermofax FP and ferrogallate architectural print (1911) copies are different again. In the former, the paper and the image both become lighter and less brown at about the same rate, and in the latter the brown paper bleaches (lighter, less brown) while the ink darkens and loses chroma (less yellowed).

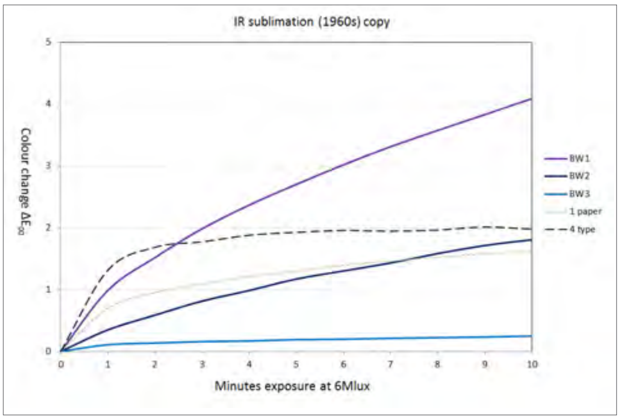

Figure 7. Fading curve of the IR sublimation copy. The type (image) colour change plateaus, while the colour change continues to climb.

It is impossible to explain the diversity in their responses; perhaps their chemical environment determines whether photoreduction or photooxidation dominates, or the image forming species were never, or no longer remain, iron (III) pyrogallic acid coordination complexes. Iron gall inks themselves are very diverse and Tse[3] has reported occasional darkening of these inks under accelerated light exposure and avoids the term "fading" to describe their response for that reason.

There is a further twist to what should be a simple story of iron gallate "fading". While the rates of colour change under accelerated conditions are consistent with published figures for iron gall inks (Reissland & Cowan 2002, Tse et al. 2010), the published figures are wrong, or rather they are an artefact of accelerated testing. An unpublished observation by Neevel[4] that like Prussian blue, their colour spontaneously reverts post-exposure (e.g. Figure 8) has recently been confirmed by Ford (2014) in a study of over 100 historic inks. Whether this happens with the ferric (pyro)gallate based copying processes (or at least the ones that become lighter with exposure) is not known, but there is clearly a possibility that they may fade more slowly in practice than indicated by the microfade testing results.

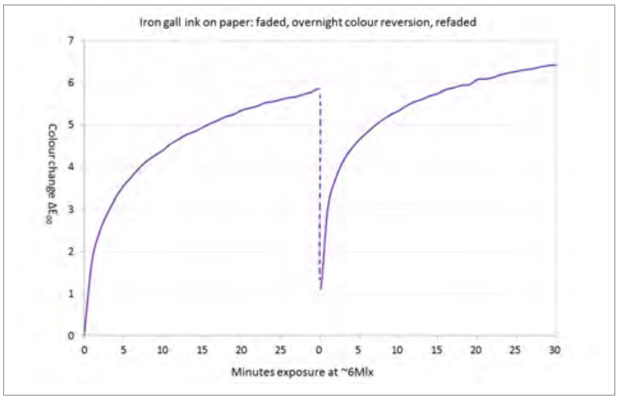

Figure 8. Experiment demonstrating overnight colour reversion of an historic (1849) iron gall ink on paper. The dashed line represents 16 hours darkness during which time much of the colour lost to the initial accelerated fading has returned.

SILVER COPIES

With the exception of the Thermofax BP (back printing) these documents are more lightfast than the dye based copies, particularly the two where the image darkened with exposure. This is not unexpected because like conventional silver-gelatine prints, which are very lightfast if well fixed, the image is theoretically metallic silver.

The Kodak thermal silver (1961) copy is the most straightforwardly lightfast, with both the paper and image darkening and becoming less yellow (blacker) at a rate equivalent to about BW3. In contrast, the image areas of the Thermofax BP (back process), which appears to be based on substantially the same principle (silver behenate soap and methyl gallate as reducing agent), became lighter and yellowed with exposure at a much faster rate (BW1-BW2). Both the fading of the Thermofax and the difference between the two are difficult to explain if the images are both composed of metallic silver.

It is logical to expect that because the coated papers are likely to contain residual silver salts and potential developing (reducing) agents they might darken on exposure, however where this was observed, the rates were comparatively low, at least while the documents were under illumination. The Thermofax BP paper had a complex response, first becoming less and then more yellow as exposure continued (Figure 9). The diffusion transfer paper (n.d.) paper did exactly the opposite, first yellowing and then bleaching, although in this case the image itself darkened. Because the diffusion transfer process relied on subsequent washing of the paper to properly fix the image if a more permanent record were desired, it should be expected that individual examples might behave differently. The dual spectrum (1967) paper was yellowed at the outer edges and whiter towards the centre of the page, indicating that something affected it during storage. The yellowed areas bleached quite rapidly (BW1-BW2) however after an initial bleaching, the whiter paper yellowed at a relatively modest rate (BW2-BW3).

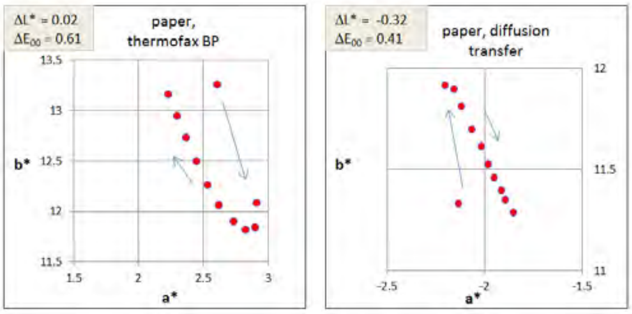

Figure 9. a* b* diagram showing direction of colour change (arrows) for the Thermofax BP and diffusion transfer copies. The former initially loses yellowness rapidly (b* decrease) and then re-yellows (b* increase), the latter responds in the opposite manner and darkens (L* negative)

CONCLUSION

Dye based copies are quite consistently fugitive to light, particularly the ubiquitous "aniline" purple typewriter "carbon" copies, spirit copy inks and official stamps still in use. Like most generalisations, however, it is not always true; much more lightfast examples of similar appearance do exist. When display limitations usually considered appropriate for such unstable colourants is a problem, individual fade testing is the only way to find out if the colourant really is unstable. Although not part of this study, some modern ballpoint and marker pen inks (including expensive brand-name examples) are similarly fugitive because they contain the same or similar dyes, however in this case it is completely impossible to generalise because inks of identical appearance many just as often be lightfast. Copy processes that utilise coated and chemically impregnated papers are intrinsically complex chemical and physical environments that are likely to undergo deterioration on several fronts, light being just one factor. Accelerated light ageing only estimates the effect of photochemical processes that occur during illumination, and while it can usually indicate where light-fading is a serious potential problem, appearance changes may also arise from other causes. Ferric ferrocyanide pigments and iron gall inks, for example, are unusual but culturally important examples of spontaneously reversible fading while, like some colour photographs, some direct thermal and other copies also undergo "dark fading".

ENDNOTES

1. Ford, B 2013, Source data compiled by Bruce Ford (2013) from various unpublished microfade testing reports for the National Museum of Australia, Australian National Archives and Museum of Australian Democracy.
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MATERIALS

Oriel Fading Test System, Model 80190
Newport Corporation
www.newport.com/Fading-Test-System/378064/1033/info.aspx

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