Australian adhesives for paper, 1870-1920

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ABSTRACT

From 1870–1920 adhesives for use with paper were either commercial preparations or made by hand in homes, studios and offices. For this research, recipes and commercial products in use in Australia during 1870–1920 were identified. However, the limited number of recipes discovered precludes any firm conclusions regarding the likely composition of unknown adhesives from this period. There is also little available information about Australian commercial products, such as Clag or Angus & Co Mucilage. It is likely that Australian usage trends mirrored those of England and America quite closely, as recipes published in international trade and amateur journals were available in Australia within months of publication. Additionally, commercial adhesives were shipped to Australia and sold by local businesses, such as those produced by the Russia Cement Company, the Leadenhall Press, the Gloy Manufacturing Company and Chas. M Higgins & Co. This paper presents the formulations and products known to be in use and the implications of these findings for the practicing paper conservator.

KEYWORDS

adhesive, Australia, conservation, glue, history, paper, starch

INTRODUCTION

Old adhesives are an integral part of a paper conservator's work, as they are often disfiguring and damaging to paper-based collections. Conservators may remove old animal glue, starch paste and rubber cement from art works, documents and ephemera in order to release collection items from old mounts, to flatten cockling or in order to attempt stain reduction treatment.

The information presented in this paper is based on research conducted by the author for a Masters degree at the University of Melbourne[1]. The research aimed to identify adhesive recipes and commercial products used on paper from 1870–1920, both internationally and within Australia. This date range was chosen to correspond with particular collections found at the State Library of Victoria, on which the author was working[2].

Recipes and product advertisements for adhesives were found in various journals and magazines of the era, in trade literature and in newspapers. Patents were also consulted, via the online European and United States patent databases[3] and records held at the National Archives of Australia. The research was limited to resources in English.

Overall, the research found that both homemade and commercial adhesives from this period were quite varied, though usually based on natural sources. Animal glue, gelatin, fish glue, starch, flour, dextrin, and gum arabic were the most common binders in use. Plant resins (e.g. dammar and mastic), polyolefins (rubber and gutta percha), gum tragacanth, linseed oil and shellac were also used, to a lesser degree. Egg albumen, casein, blood, soluble silicates and adhesives based on lime (calcium carbonate) and sugar were used, but much less frequently. Formulations containing cellulose, in which it was most likely converted to cellulose nitrate, were used on occasion. Other synthetic and semi-synthetic polymers did not come into use commercially as adhesive materials until after the 1920s.

Recipes frequently contained more than the two basic components required to make an adhesive — binder and solvent — with the addition of extra ingredients to alter various properties. Supplementary ingredients were added to increase tack, improve flexibility, extend shelf life, repel insects and to make the adhesive more resistant to water. Table 1 shows the most common additives (excluding the binders themselves) contained within all of the recipes identified, international and Australian. The role of specific additives within recipes of Australian origin will be discussed further in following sections.

Table 1: The 20 most common additives, excluding binders, in historic recipes 1870–1920, international and Australian in origin.

Ingredient	Number/percentage of recipes in which ingredient occurs	Probable uses
Water	621 (89.4%)	Solvent/diluent
Alcohol	109 (15.7%)	Solvent/diluent
Carbolic acid	100 (14.4%)	Preservative
Glycerine	73 (10.5%)	Plasticiser
Alum (aluminium potassium sulphate)	65 (9.4%)	Hardener, tackifier
Acetic acid	61 (8.8%)	Solvent/diluent
Nitric acid	46 (6.6%)	Solvent/diluent
Clove oil	42 (6.0%)	Preservative
Borax	42 (6.0%)	Solvent/diluent, hardener, tackifier, preservative
Turpentine	42 (6.0%)	Solvent/diluent, tackifier
Ammonium hydroxide	27 (3.9%)	Solvent/diluent
Essential oil (e.g. lavender)	17 (2.4%)	Preservative, flavour, scent
Sodium hydroxide	17 (2.4%)	Solvent/diluent
Aluminium sulphate	14 (2.0%)	Hardener, tackifier
Sulphuric acid	14 (2.0%)	Solvent/diluent, preservative
Calcium hydroxide	13 (1.9%)	Solvent/diluent
Mercury dichloride	12 (1.7%)	Preservative
Ammonium carbonate	12 (1.7%)	pH adjustment
Potassium dichromate	10 (1.4%)	Hardener
Lead acetate	10 (1.4%)	Colourant, hardener

RECIPES

The research identified 695 recipes for adhesives overall. Of these, only 47 recipes were found in Australian sources. These were all found in Australian photographic journals and in the 'home hints' section of some newspapers, accessed via the online Australian Newspapers database[4] hosted by the National Library of Australia.

The small number of recipes identified of Australian origin precludes any meaningful statistical analysis of the ingredients. Additionally, some of these recipes were reprinted from international sources. For example, the *Australian Photographic Journal* ran an article on photographic mountants (Standage 1902b) featuring a number of recipes provided by H C Standage, an English author who had published books on adhesives and other art materials (see Standage 1893, 1902a).

Many of the remaining recipes were located in the 'home hints' section of newspapers. They were generally simple recipes, made from easily available household ingredients. For example:

> "Zena" (St Kilda) says that a sticking paste which will not leave marks on any material which it may touch is made by pouring boiling water directly onto dry flour. The paste thus made is inclined to be lumpy, but with vigorous beating a good deal of this drawback can be overcome. The feature of the paste is that it is much stronger than other pastes, and leaves no stain. (Anon 1920)

The focus of such recipes was often to produce an adhesive that was cheap and quick to prepare, using starchy vegetables readily available in the kitchen, such as potato and onion.

> A quickly-made glue which is very strong is made by rubbing a little piece of cold boiled potato on a sheet of paper with the fingers. (Anon 1914)

A very convenient mucilage can be made out of onion juice. A good-sized Spanish onion, after being boiled for a short time, will yield on being pressed quite a large quantity of very adhesive fluid. It is a cheap and good mucilage, and answers as well as the more costly cements. (Anon 1897)

Some recipes specified use for particular purposes. For example, many were designed for the application of paper labels to glass, metal and ceramic vessels — handy adhesives to have around the house. Strength and good tack appeared to be important properties of such adhesives, achieved through the use of multiple binders or by making them up to a thick consistency. In one of the following recipes, starch and gum arabic are combined with sugar, itself capable of adhesive properties though also added for flexibility. In the other, animal glue and flour are combined to create a strong adhesive.

> A cement which will adhere perfectly to glazed surfaces, repair broken minerals, or, in fact, stick to anything, is made by taking two ounces of clear gum arable [sic], one and one-half ounces of fine starch, one half ounce of fine sugar. Pulverise the gum arabic, and dissolve it in as much water as the laundress would use for the quantity of starch indicated. Dissolve the starch and sugar in the gum solution. Then cook the mixture in a vessel suspended in boiling water until the starch becomes clear. The cement should be as thick as tar, and kept so. It can be kept from spoiling by dropping in a lump of gum camphor or a little oil of cloves. (Anon 1917b)

> A good paste for labels, suitable for bottles, may be made by soaking good glue in strong vinegar, then heat to boiling and add flour. This is very adhesive, and will not decompose when kept in wide-mouthed bottles. (Anon 1883)

The mixture of protein and polysaccharide was quite common and can be found within recipes for all sorts of purposes. This recipe was designed for 'office use':

> "Greta" (Melbourne) sends directions for making a good paste for office use. Soften by soaking in a little water 1 1/2 oz gelatine. Put it into a saucepan with one pint of water. When hot add 2 1/2 tablespoonfuls of arrowroot, mixed to a thin paste with water, and boil for five minutes, stirring all the time. Take off the stove, and stir in two teaspoonfuls of oil of cloves. This paste will keep for a very long time. I have some which was made nine months ago, and it is still good. (Anon 1918)

These recipes also indicate that resistance to decomposition was also desirable in an adhesive. This was achieved through the use of preservatives such as vinegar, camphor or oil of cloves. Carbolic acid and, on occasion, sulphate of quinine were also suggested for this purpose:

> Solutions of gum arabic soon mould and sour, and finally lose their adhesive property. It is said that sulphate of quinine will prevent this, while it imparts no bad odour of its own. The addition of a solution of a few crystals of this salt to gum arabic will prevent the formation of mould quite as effectually as carbolic acid, and by analogy it is safe to suppose that the same salt could be used in writing ink, mucilage, and possibly glue (Anon 1872)

Animal glue was a common adhesive during these years but had the disadvantage of requiring heat in order for it to be liquid enough to use. It was often cooked with an acid such as vinegar or nitric acid to create an adhesive that remained liquid at room temperature.

> A liquid cement which is useful for many purposes can be prepared as follows:-Place equal weights of carpenter's glue and vinegar in a jar, and stand it in a saucepan of boiling water until the glue has melted. Add half as much alcohol as vinegar, and, stir the cement until it is well mixed. Keep it tightly corked in a bottle. (Anon 1917c)

> This is very efficient and cheap. Boil together for several hours: Ordinary glue, 2 ounces; water, 5 ounces; nitric acid, 2 drachms. The resulting liquid is well adapted for wood, iron or steel. (Anon 1900)

Rubber-based adhesives were sometimes recommended in photographic journals, to avoid the cockling and distortion often caused by water-based adhesives. Some authors, at various times, also felt that water-based adhesives caused photographic prints to discolour and fade. Rubber-based recipes generally employed an organic solvent:

> India rubber solution is made by dissolving good india rubber in chloroform or benzine, the latter is the cheapest, but has a pungent odour. Proportions: Eight to ten grains of rubber per ounce of spirit. (Anon1902)

Gutta percha, a polyisoprene similar to rubber, was used in sheet form as a heat-set adhesive for photographic prints:

> …gutta percha tissue, an article in sheet form, on sale at all rubber supply stores and much used by tailors. It melts at a low temperature, so that a moderately heated flat-iron is the only essential to its application. It is most useful as a mounting medium. A piece the size of the print is cut, placed between print and mount, a protecting slip of paper over all, and the flat-iron is applied. The adhesion takes place at once, and with a bit of cloth any surplus may be easily rubbed from the edges of the print. (Callaway 1907)

Some recipes used alcohol as a solvent to avoid the cockling associated with water-soluble binders, though generally the binder had to be solubilised first in water. Glycerine was most likely added to the following recipe to make a more flexible adhesive — sugar was also often used for this purpose.

> Non-cockling paste: Nelson's No. 1 Gelatine, 4 ozs.; water, 16 ozs.; Glycerine, 1 oz., alcohol, 5 ozs. Dissolve the gelatin in the water, then add the glycerine and then the alcohol. (Anon 1908)

Some binders, such as shellac and mastic (both terpenes), required organic solvents for dissolution and were therefore used to create adhesives thought to prevent paper and card from cockling:

> When prints are mounted on thin cards, it too often happens that the cards "buckle" all out of shape. The following mountant

or paste will prevent it: A: White shellac, 1 oz.; alcohol (spirits), 2 oz. B: Gum mastic cut in chloroform. Add a small proportion of A to B, and apply to the print, allowing it to "set" just long enough to become a trifle "sticky" before placing the print on the mount. (Anon 1907)

Photographic journals were a rich source of adhesive recipes, partly because photography was a relatively new technology and a very popular amateur pastime. Mounting prints was an integral part of their presentation and a frequent source of grief for the amateur.

As mentioned previously, Australians undoubtedly had ready access to formulas and recipes published overseas. International photographic journals and books were often available in Australia within months of their publication, as evidenced by the acquisition date stamps on material contained in public collections. It is quite likely, therefore, that adhesives prepared by Australians could have been based on any published recipe. Overall, these recipes show that adhesives for paper during this era were varied and could contain many more ingredients than just a binder and solvent.

COMMERCIAL READY-MADE ADHESIVES

Many still-familiar adhesive trade names were developed during the nineteenth century. Higgins, Perkins, LePage and Australia's own 'Clag' were all nineteenth century tradenames, although the formulation of these adhesives has no doubt changed drastically since their original conception.

Internationally, advertisements for commercial products and tradenames were found in photographic journals, trade bookbinding literature and other publications produced for specific markets. Advertisements in more general publications, such as newspapers and magazines, were rarer, possibly because the potential market did not warrant the cost. Interestingly, adhesives were not commonly referred to in the art journals examined, except in the most general of terms. Whereas photographic journals were very much 'hands on' and 'how to', art journals of the period did not appear to concern themselves with the mechanics of mounting (for example) to any great degree. However, advertisements for adhesives have been found in journals as wide-ranging as *Century Magazine, National Geographic, and The Ladies' Home Journal.* Commercial products were also often mentioned by name in 'question and answer' columns and editorialised 'how-to' sections of newspapers and journals.

In America, trade cards were a popular form of advertising from about 1870–1900; they were less common in England and its colonies, although not unknown. These were postcard-sized advertisements featuring products and suppliers, often depicting comical scenes (see Figure 1).

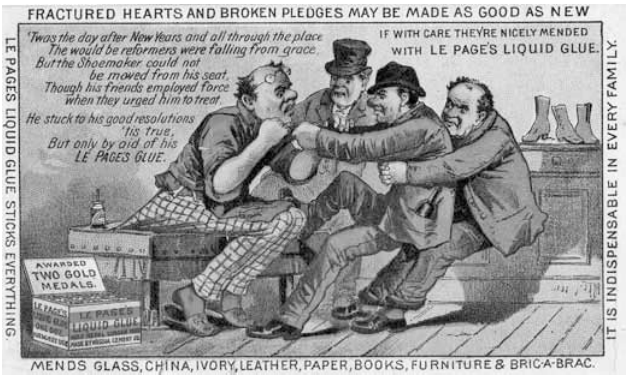

Figure 1: An example of an American tradecard produced for LePage's glue. Dimensions: 81 x 137mm. Date unknown. Collection of A. Cannon.

Trade cards were handed out in shops and at exhibitions, or included with mail-ordered goods. They were frequently collected and pasted into scrapbooks. There were two main types of trade card: stock cards, with generic images and spaces or blank backs on which messages could be printed, and cards specially commissioned by a particular company. Some European trade cards have been found in Australia in large numbers suggesting these goods were available locally; others are American in origin with local retailers' details added (O'Neill 2007, pp.13–25). English advertisers favoured magazine 'inserts', gummed or bound inside the covers of journals (Hudson 2008). For example, The Leadenhall Press frequently attached inserts for Stickphast Paste inside its publications (e.g. May 1896, see Figure 2). Collectors have not found any such Australian inserts to date (O'Neill 2007, pp.25).

Figure 2: An advertising insert for Stickphast paste. Dimensions: 221 x 141mm. Date unknown. Collection of A. Cannon.

Evidence of commercial Australian-made adhesives was hard to find. The National Archives of Australia holds the trademark application for 'Clag' (Angus 1898–99) and bottles of mucilage manufactured by Angus & Co (likely the same Angus who went on to produce Clag) have been found in antique shops. However, advertisements for such local tradenames have not been located. Sale notices in newspapers occasionally made reference to local products, such as the 1919 auction sale of the Star Glass Bottle Works (Anon 1919), which offered bottles made to contain Clag amongst other products for sale. (Whether the bottles were empty or full at the point of sale is unclear). Local glue factories were in existence, but it is also likely that many overseas products were imported.

In Australia, some businesses acted as distributors for international products, such as Harringtons Photographic Supplies and Baker & Rouse. Both businesses had shopfronts in Sydney, Brisbane, Adelaide and Melbourne. Advertisements run by these businesses and shipping records indicate that a number of American and

English adhesives were imported to Australia. Higgins' Mountant and Le Page's Clarified Glue were imported into Australia by Harrington (Harrington 1898a, 1898b); Baker & Rouse imported Kodak Adhesive Tissue (Baker & Rouse 1906a, 1906b). 'Gloy' was listed for sale by an Australian auction house on at least one occasion (e.g. Anon 1898) and was registered by trademark in various Australian colonies (Gloy Manufacturing Co. 1897a-c). Shipping arrivals list cartons and crates of such items as 'glue', 'gelatin', 'starch' and 'stationery' as well as potential raw ingredients, such as Colman's Starch (e.g. Anon 1886; 1895c), which would otherwise have been used for laundry purposes. Occasionally tradenames such as 'Stickphast', manufactured by the Leadenhall Press, were listed in ship manifests (Anon 1890). Businesses such as druggists, stationers and general stores were also likely to have sold adhesives and their raw ingredients — for example, an advertisement run by Elliot Bros. & Co (druggists and drysalters with outlets in Brisbane and Sydney) lists for sale both Nelson's and Swinbourne's gelatin and isinglass, 'Brazil' isinglass and 'Glue, Russian' (Elliot Bros. 1879a; 1879b). Some of these products were also referred to in Australian photographic journals — particularly Higgins' paste or 'Photo-Mounter' (Anon 1904; Fisher 1903; Smith 1905; Swan 1905). Le Page's (Anon 1901) and 'Seccotine' (Fisher 1903) were also mentioned.

The precise formulation of such commercial adhesives can be difficult to determine, unless associated patent records can be found. Even then, patent specifications were often vague — intentionally or not — or included statements announcing the right of the patent holder to change the formulation or to substitute ingredients as they saw fit. Le Page products from the period were often advertised as based on fish glue, but no patents relating to these adhesives have been found. Patents pertaining to Higgins products reveal them to be initially made from starch; later they employed white or yellow dextrin with the addition of antiseptics and flavourings.

Trade authors occasionally mentioned details of the composition of some commercial adhesives — for example, 'Gloy' was reported to be comprised of dextrin mixed with magnesium chloride (Rideal 1920, p.33; Standage 1902, p.90). Magnesium chloride is sometimes used as a coagulant in cooking; it may also have had this effect on the dextrin adhesive, making the product thicker and more viscous. 'Stickphast' was thought to be made from a mixture of wheat starch, plant resin, salicylic acid and oil of turpentine (Standage, 1893, p.123). Salicylic acid was most likely added as a preservative, the plant resin to increase tack and the turpentine as either a solvent for the plant resin or as another way to increase the adhesive's strength.

As part of the Masters research some analytical testing was performed on various historic adhesive samples, including a sample of Clag and Angus & Co Mucilage (see Figures 3 & 4)

Figure 3: A bottle of Clag. Date unknown. Collection of A. Cannon.

These were the only two Australian commercial products conclusively identified from the time period of interest, though a Sands & MacDougall publication (1885-1886) did feature an advertisement for 'Stickfast' (see Figure 5). This may or may not have been the same product as the English 'Stickphast', manufactured by the Leadenhall Press. An actual sample of either adhesive could not be obtained. The precise date of production of the two samples used for testing is unknown.

Figure 4: Bottles of Angus & Co Mucilage. Date unknown. Collection of A. Cannon.

The testing program included solubility and chemical spot tests, X-Ray Fluorescence (XRF) and Fourier Transform Infra-Red (FTIR). Though the results were not conclusive, they indicated that both adhesives were polysaccharide-based. Both tested positive for starch using the iodine test[5] and the FTIR spectra[6] of both adhesives contained peaks at around 3400cm^{-1} and 1000-1200cm^{-1}, peaks indicative of polysaccharides (Derrick et. al. 1999). However, the FTIR spectra for these two samples did not resemble that of a reference flour or starch spectrum, with many other peaks present. This suggests a combination of materials, whether added by design or formed by deterioration of the original product. Simple solubility tests found that, after 20 minutes, the Angus Mucilage sample was completely dissolved whereas the Clag sample was only slightly swelled. Taking into account the other results, this suggests either gum arabic or dextrin as a base for the Angus Mucilage (both are easily soluble in cold water) and starch or flour for the Clag. Commercially dextrin is perhaps a more likely base for Angus Mucilage, as it was much cheaper than gum arabic.

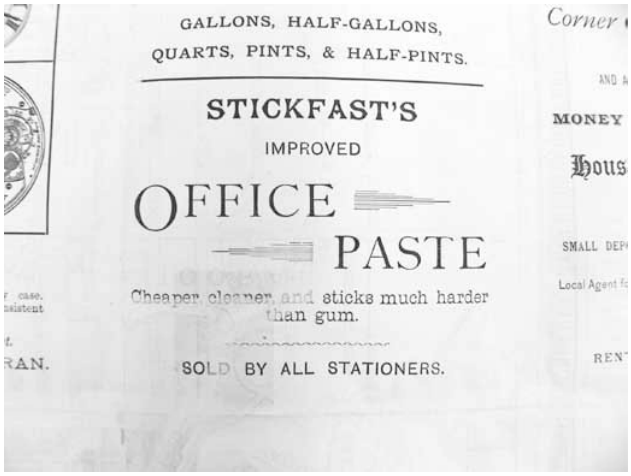

Figure 5: An advertisement for 'Stickfast paste' from a Sands & McDougall publication (1885-1886). Collection of the State Library of Victoria.

XRF[7] detected no strong elemental peaks in the sample of Clag; however the Angus Mucilage sample gave peaks for calcium, potassium and iron. Calcium and potassium are found naturally in gum arabic, as salts of arabic acid, so these results may indicate the presence of this binder. Again, however, these elements may have also leached from the glass storage bottle or metal screw-top lid or have been due to the inclusion of some other ingredient.

Space limitations preclude a detailed discussion on the issues surrounding binder identification methods here. However, further analysis of known samples may help to build a more useful reference library for identification of unknowns. Exploration of analytical techniques that are more successful with mixtures of materials would also be beneficial.

IMPLICATIONS FOR PAPER CONSERVATORS

Though there is still much unknown about adhesives from this time period, it is clear that their composition could be quite varied. As well as the usual suspects of animal glue, gelatin, starch and flour, conservators may also encounter plant resins, casein, egg albumen, rubber, gutta percha, shellac, linseed oil, soluble silicates and various mixtures of any of these materials. Old adhesives are quite likely to contain other ingredients such as metallic salts, organic solvents, acids, alkalis, essential oils and other materials added to alter the properties of the adhesive.

It is not clear how all of these organic molecules interact with each other chemically in mixtures, nor the precise effects of these reactions on the solubility of an adhesive. Combining two different binders will no doubt change the solubility of the resulting dried adhesive — for example, it could be assumed that mixing starch into animal glue will produce an adhesive with water solubility somewhere between either of those two substances alone. A variety of chemical reactions may occur between proteins, polysaccharides and other organic molecules, such as the process known as the Maillard reaction. Here protein and carbohydrate molecules combine in a condensation reaction, resulting in a new, larger molecule but with the concurrent loss of a smaller molecule, often water (Karpowicz 1981).

Multivalent metals, such as aluminium, copper and iron, are known to cause protein chains to crosslink, raising the melting point of a protein-based adhesive. This effect is also known to occur with polysaccharides (Horie 1992). However, in some cases ions such as copper (II) and iron (II) can catalyse the formation of hydrogen peroxide within proteins, resulting in depolymerisation instead of crosslinking (Banik 1989).

Aldehydes (e.g. formaldehyde) also oxidise protein and starch-based adhesives. In fact, any reagent that is reactive with amino groups, such as organic acid chlorides and anhydrides, has the potential to react with proteins and starches in this way. The mechanisms through which this occurs are not fully understood. For starch, it is likely that the aldehyde reacts with hydroxyl groups on the starch chain, resulting in the formation of bonds between methylene groups on the aldehyde and hydroxyl groups on the starch, causing crosslinking and reducing the ability of the starch to swell in hot water. It is possible that any 'polyhydroxy' acid (those having three or more hydroxyl functional groups) could have this effect. Borax (sodium tetraborate) was often used to increase the tack of starch and dextrin adhesives; phosphoric acid is also reported to increase the viscosity of potato starch (Kerr 1950, pp.190, 466-470; Kruger & Lacourse 1990).

A more in-depth discussion of the variety of reactions that may have taken place within an aged adhesive is a topic for another paper. However, this brief discussion indicates that conservators may encounter adhesives from this period that are resistant to normal water-based treatment methods. New methods of applying enzymes and chelating agents may need to be explored in order to solubilise such adhesives, such as those pioneered by Richard Wolbers (see Wolbers 2000) for cleaning oil paintings and furniture surfaces.

CONCLUSIONS

This paper has shown that adhesives from the period 1870–1920 were varied, though generally based on organic substances. Though Australians made their own adhesives, both at home and commercially, it appears that international brands were imported from overseas and therefore may also be found on Australian collections. Evidence of the precise composition of Australian commercial products is hard to locate; 'Clag' and 'Angus & Co Mucilage' are the only two commercial Australian adhesives identified from this time period. It is expected that Australian use of adhesives followed European and North American trends quite closely, as published reference material from overseas was available in Australia within months of publication. The Australian paper conservator may therefore expect to find any and all adhesive mixtures on collection material. Some of these adhesives, through design or through ageing, may no longer be easily soluble in water. Enzymes and chelating agents warrant further investigation as treatment methods.

ENDNOTES

[1] *Adhesives for paper 1870-1920: Recipes and proprietary products.* Master of Arts by Research. Department of History, Centre for Cultural Materials Conservation, University of Melbourne. Submitted August 2009. Unpublished.

[2] The Dyer Collection of land auction plans and the Victorian Patent Office Copyright Collection (H96.160/1-2766), both containing items adhered into scrapbooks.

[3] The database of the European Patent Office (http://ep.espacenet.com/) and the United States patent and Trademark Office (http://www.uspto.gov). The European Patent Office database includes British patents. The US patent database can only be searched by Issue Date, Patent Number and US classification for records prior to 1976, which limits its usefulness.

[4] The Australian Newspaper Database can be found at http://newspapers.nla.gov.au/.

[5] The test for starch using iodine/potassium iodide is a standard chemical spot test. Details can be found in the publication by Odegaard (2005; pp. 128-129).

[6] For this research, samples were ground and pressed into potassium bromide discs. The discs were analysed at CSIRO using a Perkin Elmer Spectrum 100 FTIR Spectrometer with a lithium tantalum detector4. The graphs produced were baseline corrected and normalised. Beam intensity 5912, 32 scans (summed and averaged), range 4000–400cm^{-1}, resolution 4cm^{-1}.

[7] The machine used was a Bruker AXS TRACeR® III–V handheld instrument loaned from KeyMaster Technologies. Adhesive samples were placed in the beam and analysed for 180 seconds, using a titanium filter, 15–20 kV, 15–20 micro amps and the vacuum attachment. These settings were used to reduce backscatter from the organic material and to eliminate Rhodium lines emitted by the x-ray tube. These settings allow for better detection of magnesium, aluminium, silicon, chlorine, potassium, calcium chromium and iron in organic samples (Bruker n.d.).

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