# **Cellulose Acetate Lamination: History**

#### Introduction

Lamination adds physical strength to an object by adhering one material to another stronger material. Lamination has been, and is currently, used in paper conservation / preservation to support documents using a variety of materials and adhesive technologies. This article will focus specifically on the history of cellulose acetate (CA) lamination, applied using a combination of heat and pressure. The technique was in broad use in libraries and archives in the United States and around the world from the 1930s-1990s. In some places it continues to be used (McGath et al. 2015).

One purpose of this article is to clarify some areas of potential confusion. One such area is that the term "Barrow lamination" has been incorrectly used as a synonym for all CA lamination. Barrow lamination was, in fact, only one of the many different types of CA lamination treatments that were used.

While today, CA films and plastics are known to degrade, sometimes drastically, it would be incorrect to assert that CA lamination was both a poorly conceived and underresearched treatment.

In fact, as a treatment for documents, CA lamination was arguably the first scientifically researched preservation / conservation treatment, and underwent testing and retesting throughout the decades it was in use (Scribner 1934; Scribner 1940; Wilson & Forshee 1959; Barrow 1965). However, it is true that CA lamination was often used as both a conservation and preservation treatment simply because it was the only treatment method available to institutions. CA lamination was first recommended as a preservation method by the National Bureau of Standards (NBS) (Scribner 1934; Barrow 1941) and then by William J. Barrow (Barrow 1939) with the idea that it would protect against mechanical damage and environmental contaminants and fluctuations as well as prevent biological deterioration.

Over the years institutions commonly sent Barrow their most important materials for preservation, regardless of the condition of those materials (Baker 1982). CA lamination was used to repair torn pages and to consolidate and protect burned pages (Bolsée 1950; Cutter 1967). It is often difficult to judge the effectiveness of this CA lamination treatment today because it was applied to deteriorating and intact paper alike.

There were two major CA lamination methods. The first CA lamination method was developed by the National Bureau of Standards, and as such we refer to it here as the NBS lamination method. The second was Barrow's lamination method, which evolved from the NBS method.

The NBS and Barrow methods both apply a combination of heat and pressure to adhere CA films to the surfaces of a document. Other CA lamination methods in use during the same time period typically involved the use of adhesives or solvent to apply CA to the document and are beyond the scope of this article (Minogue 1943; Cutter 1967).

The Barrow lamination method differs from the original NBS lamination method in three distinct ways: the deacidification of documents prior to lamination, the use of tissue paper in the lamination, and the type of laminator used to apply the cellulose acetate film to the document surface. These differences are highlighted in Table 1.

	Barrow Method	NBS Method	
Deacidification or pretreatment	Two bath deacidification developed in 1940: calcium hydroxide and calcium carbonate	No deacidification was recommended until 1959	
Use of outer support layer	Semi-transparent tissue layer *	No outer tissue recommneded until 1959	
Type of laminator	Separate oven and roller press heat the lamination materials prior to pushing them onto the roller **	Heat and pressure applied at the same time with a hydraulic press. Temperature, pressure, and time ranged from: 150 - 175° C, 300 - 2,000 psi and 3 1/2 - 30 minutes. (Scribner 1934)	

Table 1. Differences between the Barrow and NBS Lamination Methods

\*This is often referred to as Japanese tissue in the literature, but was rarely if ever actually Japanese tissue, rather it was usually just a semitransparent tissue paper of variable quality. The application of tissue paper was not universal, and might differ based on the type of document, the media on the document, or the state of the document.

\*\* The time exposed to the oven and the temperature of the oven were the only elements controlled. Pressure was dependent on the thickness of the paper treated (though this could be changed with the addition of blotter or other spacer material).

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#### **The NBS Lamination Method**

It should be stressed that the NBS was not trying to create a preservation method to treat all archive materials with the goal of making them last forever. Their experiments were designed to address the very urgent issue of preserving newspapers in libraries for more than a few weeks.

In 1928 the NBS (today the National Institute of Standards and Technology or NIST) and the Library of Congress began researching the application of cellulosic materials: cellophane, CA, and cellulose nitrate as strengthening agents for brittle paper (Gear 1965). They found that cellophane (reconstituted cellulose) was not robust enough to be useful as a laminating substance and cellulose nitrate damaged the paper.

In 1934 the NBS published a recommendation for the use of CA film in the lamination of newspaper. The recommendation did not promise long-term preservation, but stated that "the durability of impermanent newsprint can be greatly increased by protective coatings, but it is doubtful whether any known treatment will prevent its ultimate decay" (Scribner 1934).

The NBS used a hydraulic press which applied heat and pressure to a document simultaneously. These presses were purchased and used by a number of institutions, but were relatively expensive.

Because the hydraulic press applied pressure over the entire document, it was possible to trap air between the laminate film and the paper document. The formation of bubbles was a notable drawback of this method. However it was possible to laminate multiple sheets of paper at a single time by interleaving the laminate sandwiches with blotter papers.

With the founding of the National Archives (today the National Archives and Records Administration or NARA) in 1934, there was greater pressure on NBS to find treatment

methods with wide applicability to different materials and short application times, as this new institution was flooded with materials in various conditions.

In 1936, the National Archives purchased a hydraulic press for the CA lamination of brittle documents (Gear 1965) as CA lamination was found to be the only technique that the institution could depend on on for quick and reliable treatment of myriad documents (Scribner 1940). A summary of the NBS findings is shown in Table 2.

NBS recommended testing CA films for stability because of the chemical variation of CA films produced by various manufacturers with differences in the plasticizers used and in the processing and synthesis of the CA.

An accelerated aging test of 72 hours at 100°C (Scribner 1940) was used to determine stability. If films underwent little or no change in the course of this test, they were deemed suitable for use.

Notably the tests were done in "dry air" and thus did not simulate natural aging in even moderately humid environments. This is important because water is required for the hydrolysis of CA.

When CA breaks down under environmental conditions typical of most libraries, archives and museums, it does this using water from the atmosphere, hydrolyzing acetyl groups, and releasing acetic acid, a.k.a. vinegar (McGath et al. 2015). Thus, the predictive value of these accelerated aging tests is called into question because humidity factors were not considered in the early years of testing.

NBS had to address a variety of concerns in developing this new treatment. For example, in addition to stability of the treatment, NBS considered the additional volume and weight that lamination added to the documents. The increase to the thickness of the paper was minimized as the CA was forced into the pores of the paper under

NBS Requirements	CA lamination
1) The protective sheeting / adhesives used were stable.	<ul> <li>a) CA is thermoplastic and could be applied to paper with a combination of heat and pressure that circumvented the need for adhesives in the lamination process.</li> </ul>
	<ul> <li>b) CA films were found to be stable when tested with an accelerated aging test of 72 hours at 100° C (Scribner 1940).</li> </ul>
2) The process was simple and low in cost.	a) Lamination could be done in minutes.
	b) Minimal training was required to run the machines.
<ol> <li>The increase in weight and thickness of the treated paper was minimized.</li> </ol>	a) Thickness was only 0.0005 inch greater than that of the newspaper sheets because of compression.
	b) Weight was increased 2.5 times (Scribner 1934).

#### Table 2. NBS 1940 findings (Scribner)

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pressure and heat (Scribner 1940). "The thickness of the combined sheets was only 0.0005 inch greater than that of the newspaper sheets because of the compression of the combination, but the weight was increased 2.5 times" (Scribner 1934).

The lead scientist on the project, Scribner, highlighted the advantageous properties of CA lamination: the transparency of CA to UV, visible, and IR lights which are all used to analyze, view or photograph documents; the sheets were "water cleanable;" lamination was resistant to the passage of deteriorative gases (pollutants); and the speed of lamination made this a quicker treatment than silking. (Scribner 1940) (Silking was a technique to support brittle documents, using thin sheets of silk that were applied to either side of a document with an adhesive, typically wheat starch paste.)

Lamination treatment was evaluated by the NBS at its inception and continued to undergo testing and evaluation over the period of its use. From July 1, 1954 to June 30, 1957 the Paper Section of the NBS re-evaluated lamination and its effects on the preservation of documents. There were concerns over whether lamination was safe for the document, whether certain CA film compositions were better than others, whether lamination increased deterioration of the paper treated, and whether other variables such as the use of an outer layer of tissue or press type impacted the results.

The NBS recommended: a) specific quality specifications for CA composition, b) that alkaline pretreatment of documents (as Barrow advocated) was necessary when the paper to be laminated contained acid (especially if the lamination was done using high temperature), c) that the addition of tissue to the lamination increased the strength of the laminate, d) that the lamination showed little impact on the paper if the paper was neutral or alkaline, e) that either the flat bed or cylindrical press might be used for lamination, and f) newer non-CA plastic films might be used in lamination but future tests were needed to look at delamination, adhesion, and aging qualities (Wilson & Forshee 1959). Institutions using the NBS original method varied in their response to these recommendations.

#### The Barrow Method

William J. Barrow, a major propagator of lamination as a treatment for paper, began to treat and research decaying paper in 1932 when he founded the Barrow Restoration Laboratory at the State Library of Virginia (Roggia 1999). Barrow worked with leading preservation and conservation organizations in the District of Columbia, and transferred his knowledge and work to numerous state archives, historical societies, and libraries both in the US and abroad throughout his career (Roggia 1999; Church 2005).

From the NBS's earliest uses of lamination Barrow was interested in its effects on documents and the potential

of lamination to improve the longevity of paper and materials within archive and library collections. In early correspondence about lamination, Barrow was concerned with how lamination might be used to prevent mold or other biologically induced deterioration, in addition to lending strength to fragile paper.

Barrow began to laminate documents in 1937, when he invented his roller-press laminator which was built by the engineers of the Mariners' Museum in Newport News, VA. He sold his roller-press laminators to institutions around the world, and other companies emulated his model. While Barrow built his conservation/preservation business at the State Library of Virginia, he also started the Barrow Research Laboratory at the Virginia State Historical Society. From the invention of his laminator until his death in 1967 Barrow's lamination method was adopted by many institutions around the world. His business was perpetuated after his death by his family and colleagues, until it was closed in the early 1990s (Roggia 1999).

#### Deacidification

Barrow developed a two bath deacidification treatment in 1940 to address the issue of acidic paper. Barrow tied the loss of strength in paper to the introduction of acidic alum sizing and use of cheaper and shorter paper fibers in paper manufacture (Gwinn 1981). While not the first person to tie the issues of acidity to paper deterioration, Barrow was a leader in making the information public.

Barrow showed that if the pH of the paper was below 6.0 the acid content would continue to increase. While if the paper's pH was between 6.5 and 7.5, it was "nonacidic" and would be stable (Anon 1966). While the NBS advocated lamination as a process that might prevent paper from decaying, the act of lamination was solely a mechanical treatment and acidic paper would continue to deteriorate chemically after lamination (Anon 1966). Thus, Barrow regarded deacidification as a necessary step prior to lamination if the pH of the paper was less than neutral (Barrow 1965).

By 1946, Barrow's pre-lamination treatment with calcium hydroxide and then calcium bicarbonate was widely known, but was not in general use (Evans 1946). One of the previous advantages of CA lamination was its quick turnover time, but Barrow's deacidification pretreatment added significantly to the total treatment time of a document, which was considered a serious disdvantage.

Barrow acknowledged that in evaluating the stability of the laminate, one should consider how the pH of the CA film in combination with plasticizer loss impacted folding endurance. In his 1965 article he postulated that documents that were laminated but not deacidified prior to lamination might lose as much as half of their original folding-endurance strength (Barrow 1965). Barrow goes on to state that a

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much slower rate of deterioration was associated with those documents that were deacidified prior to lamination than those laminated without deacidification. After further improvements were made to the lamination process in the mid 1940s he predicted a very slow rate of deterioration for papers that were deacidified.

#### Tissue

Almost from the inception of the Barrow lamination method, Barrow advocated for the use of "a strong, well purified cellulose fiber tissue" paper as the most external layer of the laminate to increase its tear resistance and produce a matte surface (Barrow & Carlton 1968). While the outer tissue paper was commonly referred to as Japanese tissue, what was used was often a semi-transparent tissue of variable quality.

Not all documents or document types were treated in the same way in the Barrow Lamination process. In some cases, such as for burned or darkened documents, tissue paper was not included as the external layer of the laminate. The use of tissue paper could obscure the media on the documents as was seen in the Belgium General State Archives where Barrow consulted on the lamination of documents that had fire-damage. In that case the documents laminated with tissue paper were delaminated using acetone baths and re-laminated without tissue paper (Bolsée 1950).

For documents where only one face held information or media (as with many maps) Barrow's lab used a layer of muslin to strengthen the backside of the laminate. However, many institutions did not include tissue paper in their lamination treatments, following the original NBS methods.

#### Laminator

Barrow invented the roller laminator in 1937 while at the Mariners' Museum, whose engineers built the first model (Roggia 1999). This invention started his lamination business (Marwick 1964).

The roller laminator was less expensive to purchase and use than the hydraulic press used in the NBS study. It was an improvement over the hydraulic press because it reduced the formation of bubbles in the laminate, distributing the pressure more evenly over the document. It also sealed edges more securely and relied on air cooling rather than artificial cooling (Barrow 1939).

The roller laminator was limited in the size of the documents that could fit through the laminator. This meant that oversized materials, like maps, were cut into sections prior to lamination to fit through the laminator. (This limitation would also apply to a hydraulic laminator depending on the size of the hydraulic press.)

#### **CA Lamination Films**

By 1940, there were already different CA films available from a variety of vendors, however specific vendors were not mentioned in the NBS report (Scribner 1940). While it is not known which films were used by all institutions, the histories of NARA and Barrow's research laboratory may provide some guidance to the procedures and materials used. NARA and Barrow were leaders in the study and use of lamination, so understanding the trends in what they were using can shed light on what was considered the "gold standard" by other institutions.

Protectoid was the brand of film that was used at NARA when they began lamination in 1936-37 until 1941 when they started to use DuPont's 88CA<sub>4</sub>8 and then switched to the Celanese Corporation of America's P-911 in 1957 (Gear 1965). Barrow's account books show that his shop bought CA film from both Celluloid Corp and DuPont in 1941, but by 1942 was only buying from DuPont (unpublished notes). His records seem to indicate that the Barrow shop continued to use and recommend the use of the DuPont CA film until it was discontinued in 1971(unpublished notes; Barrow 1953).

At that time an Eastman Kodak CA film was found to be suitable for lamination by Barrow's Research Laboratory (unpublished notes) and appears to have been used until the Barrow Restoration Laboratory closed in the early 1990s. The exact composition of these films is unknown to the author at this time, as little information on the compositions is available in the literature, and the compositions from individual manufacturers may have changed over time.

#### **CA Lamination Deterioration**

Barrow responded to observed deterioration in early laminate films, conducting and publishing research on early laminates. He highlights in his 1965 paper that CA films purchased between 1938-1941 (independently identified as coming from Celluloid according to Barrow's account records) were more acidic than desirable and released an acetic acid odor (Barrow 1965). He believed that this was a result of the cleavage of acetate groups due to residual sulfuric acid from the manufacture of the original CA.

In 1965, he published a paper that looked at the stability of documents that had been treated in the first years of lamination. In this paper, he stated that thousands of deteriorated documents were restored by deacidification and lamination by his shop in the period from 1938-1965. He continued that he had not seen in that time evidence of any deacidified and laminated document becoming more brittle due to deterioration. Barrow tested "reclaimed film" for acid, two samples for each year from 1938-1956, and showed that the films he used after 1941 (Dupont's films) were "relatively free of acid." He proposed that the introduction of magnesium acetate to the film by the manufacturers eliminated the acidic condition.

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#### **End of CA Lamination**

According to Jones (1987), the critiques of lamination and Barrow's methods started in the mid-1970s after a paper published by Frazer Poole of the Preservation Division of the Library of Congress that highlighted the use of encapsulation over lamination. Poole examined the issues tied to lamination: that it employed heat and pressure which could damage the paper during lamination; and that acidic paper continued to deteriorate after lamination (1976).

While these concerns were not new, encapsulation offered an alternative that was fast, did not employ heat or pressure, and was easily reversible. In the subsequent decades many institutions across the US began to encapsulate documents that would previously have been laminated, with most halting their use of lamination in the 1980s and early 1990s (McGath et al. 2015).

#### Today

The question of the long-term stability of laminated documents remains open. While a recent survey by this author showed that well over three million documents in the United States have been laminated using CA lamination methods (either NBS or Barrow), fewer than 0.6% of those documents have been delaminated.

The reasons for delamination vary but include observed deterioration, aesthetic concerns, worry over potential deterioration, or to ascertain that delamination is possible. Most CA laminated collections appear to be in relatively good condition at this time according to the survey done by the Heritage Science for Conservation group at Johns Hopkins (which will be covered more fully in a future article)(McGath et al. 2015). However, as CA ages and undergoes hydrolysis, it becomes more difficult to remove by submersion in acetone, thus, as a community we should remain vigilant.

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