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Artificial Ageing of Paper: Scientific Foundations, Methodological Framework, and Applications in Archival and Library Preservation

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Abstract:

Purpose - This paper presents the theoretical basis, methodological framework, and practical applications of the artificial ageing of paper, a key technique for predicting and improving the long-term stability of archival and library materials. It aims to familiarize conservators, archivists, and librarians with the fundamental concepts necessary for interpreting scientific literature and making informed preservation decisions.

Design/methodology/approach - The article introduces key chemical mechanisms of paper ageing, particularly acid hydrolysis and oxidation of cellulose. It presents standardized accelerated ageing methods and discusses two core indicators of paper degradation: degree of polymerization and folding endurance. Mathematical models are employed to explain ageing rates and reaction kinetics, including Arrhenius-based estimations of useful paper lifespan. Experimental setups for artificial ageing and comparative evaluations of conservation treatments are illustrated using both real and hypothetical data.

Findings - This article synthesizes well-established scientific knowledge and practical methods related to artificial ageing of paper. By organizing complex concepts into an accessible framework, it clarifies how artificial ageing can be used to assess paper stability, evaluate conservation interventions, and understand the influence of environmental conditions. The discussion reinforces widely accepted conclusions in the field, such as the impact of acidity, the role of deacidification, and the value of alkaline-buffered materials.

Originality/value - This paper is intended as both a practical reference and an educational tool. It bridges scientific methodology and conservation practice, offering a synthesis of theory, standards, and field applications. It enhances the scientific literacy of preservation professionals and supports evidence-based conservation and storage decisions. The clear mathematical modeling of degradation processes makes the content accessible even to readers without advanced technical backgrounds.

Practical implications - The synthesis presented here can guide practitioners in selecting conservation treatments, evaluating

storage materials, and establishing preservation conditions to help extend the lifespan of archival holdings.

Research limitations/implications - While accelerated ageing simulates natural degradation, extrapolations to long-term behavior must be made cautiously due to potential non-linearities and model assumptions.

Index Terms — Artificial ageing; Ageing kinetics; Paper degradation; Arrhenius equation; Archival storage; Deacidification.

I. INTRODUCTION

This article presents the theoretical background, mathematical framework, and practical applications of artificial paper ageing in the conservation and preservation of archival materials and books.

Key applications of accelerated ageing as a methodological tool include [1-3]:

- Investigating factors influencing paper ageing rates, with a focus on temperature, relative humidity, and pH.
- Investigating the mechanisms and the kinetics of paper deterioration.
- Evaluating conservation methods and materials used in paper preservation.
- Determining optimal storage conditions for archival and library materials.
- Estimating the useful lifespan of various types of paper.
- Developing paper with enhanced chemical and mechanical stability, suitable for long-term preservation (permanent paper).
- Researching and selecting appropriate materials and methods for archival storage, including archival papers, cardboard, and adhesives.

Understanding the principles and rationale behind the development of archival and library storage and preservation standards enables professional archivists and librarians to fully appreciate their importance, as well as the consequences of non-compliance. This knowledge also equips them to make informed and conscious decisions when standard guidelines and "preservation formulas" are insufficient or unavailable.

The additional applications of artificial ageing presented

in this article introduce key concepts essential for interpreting preservation and conservation literature. These concepts are directly linked to preservation and conservation management within archives and libraries. A strong grasp of these principles will help archivists and librarians effectively utilize scientific literature to protect and safeguard the written cultural heritage.

To support readers lacking formal education in chemistry or mathematics, this article offers foundational explanations, supplemented by simplified examples and the underlying mathematical principles employed in the presented calculations.

II. BASIC CONCEPTS

A. Composition of Paper

Paper is primarily composed of plant fibers that have undergone various treatments. In hand papermaking, it is formed by filtering a suspension of fibers (paper pulp) through a specially designed sieve-like screen known as mould. The fibers deposit onto the screen, forming a relatively uniform layer that, when dried, acquires cohesion and mechanical strength. [4]-[5].

The fibers are essentially plant cells with various components removed. Cellulose (Fig. 1), a natural polymer, is the main component of plant cells and is the desired raw material for making paper. Thus, the various treatments of the original vegetable matter aim to purify the cellulose to varying degrees, depending on the desired quality and yield of the final product. The best quality paper is produced from cotton, linen or chemical pulp (wood pulp with the lignin chemically removed, [4]-[5]). Newsprint is produced from wood pulp that contains a large part of the original components of plant matter (mechanical pulp, mainly produced by mechanical pulping of wood, [4]-[5]).

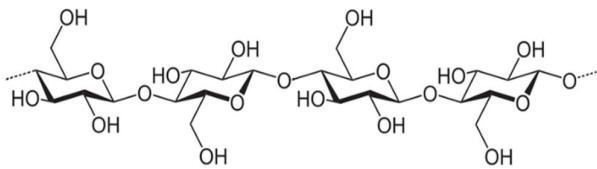


Fig. 1. The cellulose macromolecule is composed of n β -glucose ($C_6H_{12}O_6$) units linked by $1 \rightarrow 4$ β -glycosidic bonds, with the simultaneous release of $n-1$ water molecules. n equals the degree of polymerization explained below.

Cellulose is a linear polymer of β -glucose ($C_6H_{12}O_6$, Fig. 2) [4]-[6]. It consists of a large number of glucose molecules (in typical paper varying from 800 to 2,000) linked by β -(1 \rightarrow 4) glycosidic bonds (Fig. 3). As the primary component of paper, cellulose plays a crucial role in determining its mechanical strength.

In addition to cellulose, other components of paper derived from the original plant material are hemicelluloses and lignin [4]. Hemicelluloses are polymers of various sugars, similar in chemical composition to cellulose. They are compounds of relatively low molecular weight and a significant part of them remains in the paper. Lignin is also a

polymer found in high concentrations in wood, but with a completely different chemical composition and structure from cellulose. Lignin is a structural component of plants, deposited around cellulose fibers, where it acts like cement, while the cellulose fibers serve as reinforcement, similar to an iron framework in the plant's structure. Lignin is partially or completely removed from the chemical pulps, while a large part of it remains in the mechanical pulps. Due to its rigidity and hydrophobic nature, it does not allow for strong interconnections between the paper fibers. Thus, paper containing lignin has low initial mechanical strengths and, due to the chemical instability of lignin, changes color during ageing (yellowing, [7]). However, lignin does not appear to affect the mechanical permanence of papers made from mechanical pulp, as it functions as an antioxidant. [8-10].

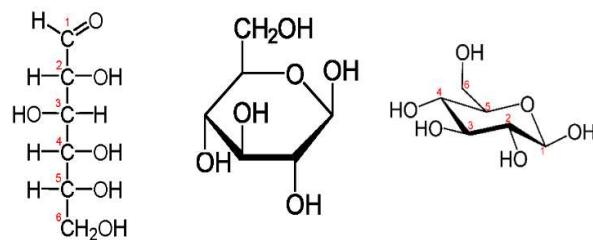


Fig. 2. Structural and stereochemical formulas of β -glucose. The designation β - indicates that the $-OH$ group on carbon 1 is oriented upwards. Cellulose is composed of β -glucose monomers linked between carbons 1 and 4 via an oxygen bridge. In the second and third formula, the carbon and single hydrogen atoms are omitted.



Fig. 3. Formation of a glycosidic bond. The glycosidic bond is formed and a water molecule is removed from the hydroxyls (-OH) of carbons 1 and 4 of two adjacent glucose molecules. The two carbons are connected by an oxygen bridge.

Paper is a hygroscopic material, meaning it absorbs or releases moisture from the surrounding air until it reaches Equilibrium Moisture Content (EMC). The relative humidity (RH) of the environment is the primary factor that determines the water content of paper, while temperature influences the rate at which moisture is absorbed or desorbed. At ambient temperatures and 50% RH, the moisture content of most common types of paper typically ranges between 5% and 7%.

B. Mechanisms of the Natural Ageing of Paper

During paper ageing, the main chemical degradation mechanism is the partial hydrolysis of the glycosidic bonds due to the presence of acids (acid hydrolysis of cellulose). Hydrolysis, as its name suggests, is the breaking of a chemical bond with the simultaneous addition of water [2]-[4]-[6]. The necessity of the presence of water explains why increasing humidity increases the rate of hydrolysis [11]. This reaction can be represented schematically with the opposite course of the reaction shown in Fig. 3. The result of partial hydrolysis is the breaking of cellulose macromolecules into

smaller pieces, which are also shorter cellulose macromolecules that contain fewer glucose molecules than the original. This reduction in the average length of cellulose macromolecules has severe consequences for paper, leading to diminished mechanical strength and flexibility, increased brittleness, and, in extreme cases, the potential disintegration of the paper sheet during use.

Hydrolysis of cellulose is catalyzed¹ by acids (acid hydrolysis) and enzymes [2]-[4]-[6]. Acids are absorbed from the environment, particularly from acidic air pollutants, introduced during paper manufacture, or produced during paper ageing [2]-[12]-[13]. Enzymes are secreted by microorganisms such as fungi and bacteria [14].

Other chemical mechanisms, such as oxidation, act synergistically to accelerate cellulose hydrolysis and, consequently, the ageing of paper. Oxidation plays a significant role in the color change (yellowing) of paper over time [7]-[15].

C. Artificial or Accelerated Ageing

The method of accelerating the ageing of a material by increasing the temperature to draw conclusions about its natural ageing at ambient temperatures is known as thermal artificial ageing or accelerated ageing. For the straightforward comparison of the ageing resistance (permanence) of various types of paper over time, the International Organization for Standardization (ISO) has standardized four thermal artificial ageing procedures, the most commonly used of which is described in ISO 5630-3 [16]. This standard prescribes exposing samples to a temperature of 80°C and 65% relative humidity for one or more time intervals. Selected paper properties (typically folding endurance or degree of polymerization) are measured before and after artificial ageing, and the retention of these properties is calculated. The greater the retained value of these properties after artificial ageing, the higher the paper's resistance to ageing.

This simple and rapid method can also reveal the impact of various treatments (such as different conservation interventions) on the ageing resistance of a paper type [1]-[17]. This method is accepted even by critics of using the Arrhenius equation (see below) to estimate the useful lifespan of paper [2]-[18]-[19]. Readers interested in the natural and artificial ageing of paper and cellulose are referred to the literature reviews on these topics by Fellers et al. [20], Emsley and Stevens [21] and Zervos [2].

D. Two Important Properties of Paper

At this point, the definitions of two key properties of paper are provided, as they are important indicators of paper degradation and will be essential for the subsequent discussion.

Degree of Polymerization (DP) of cellulose. The number of glucose molecules linked together to form a cellulose macromolecule is referred to as the Degree of

Polymerization. [4]-[6]. Cellulose macromolecules derived from natural sources exhibit varying degrees of polymerization, even in the same paper sample. Thus, an average value, the average degree of polymerization, is used to characterize a specific paper sample. The degree of polymerization for cotton is approximately 7,700 (and can reach up to 12,000), around 3,000 for wood cellulose, and between 600 and 1,600 for processed wood pulp.

The degree of polymerization of cellulose can be measured by several methods. One commonly used approach is viscometry, where the intrinsic viscosity of a cellulose solution is measured, and DP is calculated based on established relationships between viscosity and chain length [6]-[22]. Another method involves gel permeation chromatography (GPC)², which separates cellulose molecules by size, allowing for a more direct assessment of molecular weight distribution and DP [23]-[24].

The average degree of polymerization of the cellulose contained in a paper sample is related to its mechanical strength and its general quality. During both natural and accelerated ageing of paper, the average degree of polymerization decreases due to the hydrolysis mechanism, as explained earlier. If the initial degree of polymerization is DP_0 and after ageing for time t it decreases to DP_t , the fraction of the broken glycosidic bonds (δ) at the end of time t can be calculated using equation (1) [2]-[11]-[20]:

$$\delta = \frac{1}{DP_t} - \frac{1}{DP_0} \quad (1)$$

Studies have shown that during the initial stages of hydrolysis, the fraction of broken glycosidic bonds (δ) increases linearly with ageing time (2), indicating a constant rate of bond breakage [2]-[11]-[25]:

$$\delta = k \cdot t \quad (2)$$

where k is the rate constant and t the ageing time.

Monitoring the number of bonds broken during ageing provides authentic chemical kinetics data, as it is directly linked to the chemical mechanism of ageing, the cleavage of glycosidic bonds.

Folding Endurance. The decimal logarithm of the number of complete folds a paper specimen can withstand under tension before breaking (3) [20]-[26]:

$$Folding Endurance = FE = \log [number of folds] \quad (3)$$

The measurement is carried out on a strip of paper 15 mm wide and 10 cm long as shown in Fig. 4 [26]. Folding endurance is the most sensitive mechanical property for the detection of changes induced to the samples by accelerated or natural ageing, since it decreases faster than other mechanical properties of paper [1]-[2]-[17]-[27] and is directly connected to paper usability [17]-[28]. Its dependence on the ageing time at the initial stages of ageing is also linear, that is, its graph as a function of time is a straight line [2]-[29]-[30]. For these reasons it is the most commonly used mechanical property for determining the

¹Catalysis: acceleration of a reaction due to the presence of a substance, the catalyst, which is not consumed during the reaction.

²Synonym: size exclusion chromatography (SEC)

rate of paper ageing.

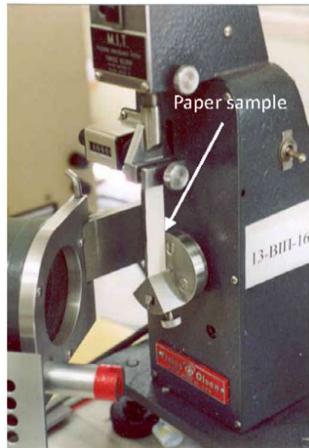


Fig. 4. The folding endurance tester of the General Chemical State Laboratory (G.C.S.L.) in Athens [MIT instrument type, [20 p. 27]]. The paper sample is held by two clamps. The lower clamp is movable and folds the sample to an angle of 135° in either direction. The desired stress is applied to the specimen by an adjustable spring.

III. DETERMINATION OF THE RATE OF PAPER AGEING

Ageing leads to the deterioration of the physical, chemical, and mechanical properties of paper. The rate of ageing is determined through experiments that monitor and record the degradation rate of these properties. Among the most commonly used indicators are the Degree of Polymerization (DP) and Folding Endurance (FE).

In the next discussion, the determination of the rate of paper ageing will be demonstrated by monitoring folding endurance³, followed by assessing the fraction of broken bonds, in two fictional experiments based on accelerated ageing of two different specific types of paper.

These fictional experiments, though based on hypothetical values for folding endurance and degree of polymerization, demonstrate a plausible approach to determining the rate of ageing for two specific types of paper at a specific temperature and relative humidity (here, 80°C and 65% RH). Both experiments take place in a ventilated⁴ laboratory oven capable of maintaining stable and uniform temperature and humidity conditions. An adequate quantity of paper samples is suspended within the oven, and at predetermined intervals a set of samples is removed from the oven and tested for the desired property.

In the first experiment, the rate of ageing is determined

³ Experiments that monitor paper properties other than the fraction of broken glycosidic bonds δ , such as mechanical or optical properties, fall under the category of "Property Kinetics" studies.

⁴ Specialized ovens that continuously renew the internal atmosphere are employed, a necessary condition for the effective removal of gaseous by-products generated during the ageing process. This is significant, as the accumulation of such products may influence the kinetics of the ageing reactions.

⁵ Folding endurance is highly dependent on the paper's water content. Preconditioning and conditioning are essential to obtaining accurate results, as they standardize the humidity absorbed by the samples, ensuring a consistent moisture level across all measurements.

by following folding endurance. Each set of samples is removed from the oven, preconditioned at 23°C and 25% relative humidity for 24 hours, and then conditioned at 23°C and 50% relative humidity for an additional 24 hours⁵. Once conditioning is complete and under the same environmental conditions, the folding endurance of each set of samples is measured.

To clarify the process, imagine that the experiment begins with 60 strips of paper. Initially, 10 strips are set aside, preconditioned, conditioned, and then tested for folding endurance, with the result recorded as the average of these 10 measurements. After 10 days of ageing, another 10 strips are removed from the oven, preconditioned, conditioned, and measured for folding endurance. This process is repeated every 10 days until the experiment concludes on the 50th day. The results are presented in the following Table I⁶:

Table I: Folding Endurance values against days of ageing

Days of Ageing (d)	0	10	20	30	40	50
Folding Endurance (FE)	2.81	2.75	2.69	2.66	2.61	2.54

The folding endurance values are then plotted against ageing time (Fig. 5).

The points on the graph do not align perfectly in a straight line, but they are close enough to suggest a linear relationship⁷. Using a mathematical technique called linear regression, a straight line that best fits these points can be drawn, and its equation can be calculated⁸ (4):

$$FE = -0.0051 \cdot d + 2.8052 \quad (4), \text{ } d \text{ stands for days}$$

According to equation (4), the rate of change in folding endurance (FE) equals -0.0051 per day of ageing, which can be considered the ageing rate for the specific paper tested under the conditions of the experiment. The negative sign indicates a decline in FE over the course of the experiment. This analysis is only valid if the data points align closely with a straight line, or else, fit the linear model well enough, to justify this approach.

⁶ Recall that $FE = \log [\text{number of folds}]$. If for example the sample withstands 1000 folds, then $FE = \log 1000 = 3$. The first value of FE in Table I is 2.81 and it corresponds to 646 folds ($\log 646 = 2.81$).

⁷ In real life experiments, the points may not fit the linear model so well. However, it is important to remember that this experiment is fictional, and the values are hypothetical, intended solely to illustrate the procedure.

⁸ This can easily be performed using spreadsheet software like Microsoft Excel or statistical software such as SPSS. The linear model follows the general equation $y = ax + b$, where a represents the slope of the line, indicating the rate of change in y (in this case, folding endurance, FE). The coefficient of determination, R^2 , can also be calculated effortlessly using these tools. R^2 reflects how well the data align with the regression model, demonstrating the goodness of fit. R^2 equals 1 for a perfect fit. In this experiment, $R^2=0.9904$, indicating an excellent fit to the linear model.

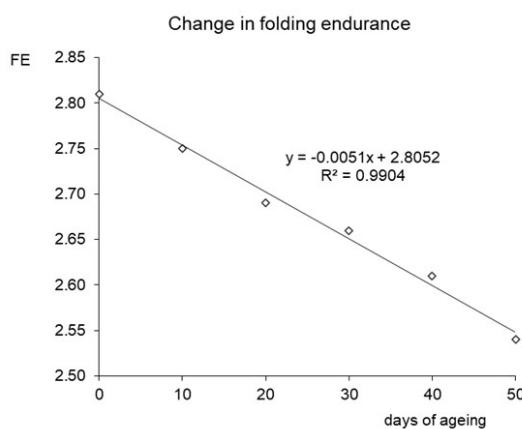


Fig. 5. Plot of the FE values against days of ageing

Here, it is assumed that the rate of the reaction k equals the rate of the reduction in the FE values, which can be calculated by the following formula (5):

$$\text{Ageing rate} = k = \frac{\text{change in FE}}{\text{time}} \quad (5)$$

A similar process applies if, instead of monitoring folding endurance, the change in the fraction of glycosidic bonds broken during ageing (δ) is observed. In this second experiment, the withdrawal of a set of two paper samples occurs every 5 days. The degree of polymerization is determined as the average DP value of the two paper samples and from this, the fraction of broken bonds (δ) is calculated using equation (1). This fictional experiment might yield the results shown in Table II.

Table II: δ values against days of ageing

Days of Ageing (d)	0	5	10	15	20	25
Fraction of broken bonds (δ)	0	0.00023	0.00037	0.00062	0.00077	0.00096

The values of the fraction of broken bonds are then plotted against ageing time (Fig. 6). As in the case of folding endurance, the points in the graph are not exactly in a straight line, but again they are very close to it, suggesting that this process is also linear. Once again, a straight line that best fits the points is drawn using linear regression, and its equation is calculated (6):

$$\delta = 0.00004 \cdot d \quad (6), d \text{ stands for days}$$

The only difference in this case is in the shape of the graph. This would be a straight line passing through zero, and the calculated rate would be positive, as more bonds break with ongoing ageing. According to equation (6), the rate of change in δ for this paper is 0.00004 bonds per day of ageing⁹.

As seen above, it is assumed again that the rate of the reaction k at 80°C and 65% relative humidity equals the rate of the reduction in the δ values, which is calculated by a similar formula (7):

$$\text{Ageing rate} = k = \frac{\text{change in } \delta}{\text{time}} \quad (7)$$

Examples of the methodology demonstrated above are widely available in the relevant literature. For instance, see Roberson [30 p. 148, fig. 1] and Feller et al. [25 p. 18, fig. 4]. Additionally, there is an extensive body of literature for those interested in exploring the natural and accelerated ageing of paper in greater detail [2]-[16]-[20]-[31-40].

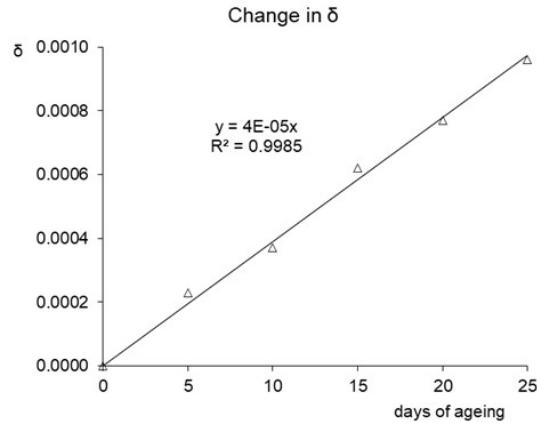


Fig. 6. Plot of δ values against days of ageing

IV. ARRHENIUS STUDIES – DETERMINATION OF THE USEFUL LIFESPAN OF PAPER

In the previous chapter, the determination of the ageing rate of paper at a relatively high temperature was demonstrated. The same methods could theoretically be applied to estimate the useful life of paper stored in libraries and archives at ambient temperatures. However, this approach poses a significant practical challenge: paper ageing at room temperature is an exceptionally slow process. Detectable changes in the properties of paper may require many years to manifest. Although such long-term experiments have been conducted [18]-[41] and others are in progress [42], a method that could provide relatively rapid and scientifically valid results is highly desirable. Such a method exists [2]-[11]-[29]-[43]-[44] and is grounded in a fundamental principle of chemistry expressed mathematically by the Arrhenius equation (8):

$$k = A \cdot e^{-\frac{E_a}{R \cdot T}} \quad (8)$$

where k is the rate constant, A is the pre-exponential factor (or Arrhenius factor or collision frequency factor), E_a is the activation energy of the reaction, R is the universal gas constant and T is absolute temperature (°K)¹⁰.

The Arrhenius equation quantifies the well-known chemical principle that reaction rates increase with rising temperature. Its importance lies in its ability to link the rate constant k of a reaction to temperature. Given the values of A and E_a , this equation enables the calculation of the reaction's rate constant at any specified temperature. Before examining how this is done, it is necessary, for

⁹ $R^2=0.9985$.

¹⁰ The constant R has a value of 8.314 J/K·mol. To convert temperature θ from degrees Celsius to Kelvin (T), the following relationship is used: T (in Kelvin) = θ (in Celsius) + 273.

reasons of scientific validity, to discuss the extent to which the above equation can be used to the study of paper ageing [11].

The Arrhenius equation applies to a specific chemical reaction, where the rate constant k represents the rate at which particular quantities of reactants are converted into products. However, the ageing of paper involves a multitude of reactions, some occurring in parallel, others in sequence, and some in competition. It has been shown that in such cases, the equation can be used under a certain condition [11], which will be discussed later.

Regarding the reaction rate, if the monitoring of δ (fraction of broken bonds) is used, there is no concern, as this approach provides authentic chemical kinetics data, as previously mentioned [45]. However, if folding endurance or another property of paper is used, the measurements do not pertain to quantities of reactants and products but rather to the change in a mechanical property of the paper [45].

On the rationale that changes in paper properties are due to the chemical alterations it undergoes during ageing, many researchers have applied the methodology based on the Arrhenius equation to the study of paper ageing by following the change in FE [30]-[43-48]. While a degree of skepticism exists within part of the scientific community regarding this logical leap [27]-[48]-[49], the methodology to be presented is widely accepted, and its results to date have shown few inconsistencies or gaps. In the following analysis, the terms "rate constant, k " and "reaction rate" are used interchangeably, even though they do not strictly align with their precise scientific meanings.

As discussed above, the problem of calculating the reaction rate at any given temperature T is reduced to determining the values of A and E_a . This calculation is accomplished through the following technique. First, equation (8) is transformed into its equivalent, equation (9), which is more convenient to use. This transformation is achieved by taking the natural logarithm¹¹ of both sides of the equation¹²:

$$\begin{aligned} k &= A \cdot e^{-\frac{E_a}{RT}} \Rightarrow \ln k = \ln(A \cdot e^{-\frac{E_a}{RT}}) \Rightarrow \\ \ln k &= \ln A + \ln e^{-\frac{E_a}{RT}} \Rightarrow \ln k = \ln A - \frac{E_a}{RT} \Rightarrow \\ \ln k &= -\frac{E_a}{R} \cdot \frac{1}{T} + \ln A \quad (9) \end{aligned}$$

Equation (9) has the following general form:

$$y = \alpha \cdot x + \beta \quad (10)$$

where $y = \ln k$, $\alpha = -\frac{E_a}{R}$, $x = \frac{1}{T}$, and $\beta = \ln A$.

The general equation $y = \alpha \cdot x + \beta$ (10) for negative α produces a straight-line graph as depicted in Fig. 7. The line intersects the y-axis at β (correspondingly, for equation (9) at the point $\ln A$) and has a slope $\tan \omega = \alpha$ [for equation (9), the slope $\tan \omega = -\frac{E_a}{R}$ (Fig. 7)].

¹¹ The natural logarithm of a number is its logarithm to the base of the mathematical constant e , which is approximately equal to 2.718. The natural logarithm of x is the power to which e should be raised to equal x .

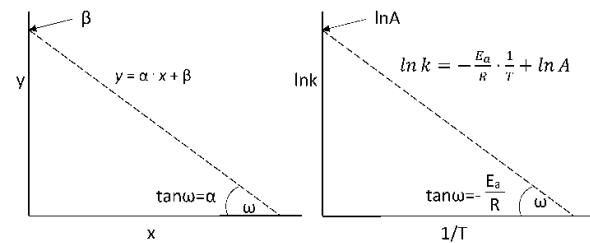


Fig. 7. Generalized plots of equation $y = \alpha \cdot x + \beta$ and Arrhenius equation.

By plotting $\ln k$ as a function of $1/T$, the value of $\ln A$ can be determined from the intercept of the graph with the y-axis, and $\frac{E_a}{R}$ (and thus E_a , since R is known) from the slope of the line. Alternatively, computational methods (linear regression analysis) can also be used, as shown below. Such a plot is called an Arrhenius plot. This can be achieved by calculating the ageing rate (i.e., the rate constant k) as shown above at various temperatures and constructing the plot of $\ln k$ as a function of $1/T$.

The results of such an experiment are presented below [Table III, equation (11) and Fig. 8]. This experiment consists of a series of tests similar to the one described in Section 3, where the rate of change in the fraction of the broken bonds (k) is measured at various temperatures using accelerated ageing. Typically, temperatures below 100°C are used to avoid triggering reactions in the paper that wouldn't occur at room temperature, and above 50°C to minimize the sample dwell time in the oven. It goes without saying that longer times are used for lower temperatures. It is crucial in these tests to maintain a constant paper water content, uniform across all temperatures, which is achieved by appropriately adjusting the relative humidity according to the oven temperature.

Table III: Rates (k) of change in the fraction of broken bonds (δ) and $\ln k$ at five different temperatures, as measured by accelerated ageing experiments according to the method shown in Chapter 3.

θ (° c)	$1/T$ (° K)	k (days ⁻¹)	$\ln k$
90	0.00275	0.00023143	-8.3721
80	0.00283	0.00008919	-9.3257
70	0.00292	0.00002197	-10.7271
60	0.00300	0.00000885	-11.6368
50	0.00310	0.00000200	-13.1237

Once the rates k at the chosen temperatures are determined, their logarithms ($\ln k$) and the inverses of the absolute temperatures ($1/T$) are calculated, and a plot of $\ln k$ as a function of $1/T$ is constructed. From this plot, the values of $\ln A$ and E_a can be determined either graphically or through computational methods (linear regression analysis). For this calculation to be valid, the plot must form a straight line, which is the condition previously mentioned as

¹² The following properties of logarithms are applied for the conversion: $\ln(a \cdot b) = \ln a + \ln b$ and $\ln(e^x) = x$

necessary for this methodology to be applicable [11].

The points in the graph (Fig. 8), although not exactly in a straight line, they are very close to it, indicating that the process is linear and that the above-mentioned methodology is applicable. A straight line that best fits the points is drawn using linear regression with the following equation (11):

$$\ln k = -13863 \cdot \frac{1}{T} + 29.847 \quad (11)$$

From equation (11), the value of $\frac{E_a}{R}$ is derived, which equals 13863, and the value of $\ln A$, which is 29.847. By solving for E_a and substituting R by its value (8.124 J/mol), E_a is calculated as 115,257 J/mol or 115.257 kJ/mol. A is derived from $\ln A$ as 9.14×10^{12} . Both values of E_a and A closely align with values reported in the literature [2]-[11]¹³.

Equation (11) can now be used to calculate the value of $\ln k$ and, from that, the value of k at any given temperature. Assuming that room temperature is 20°C, from equation (11) the rate constant k is calculated to be 2.85×10^{-8} . Comparing this to the rate constant at 80°C (which is 8.92×10^{-5} according to Table III), ageing at that temperature is 3,128 times faster, meaning that for this specific type of paper, 1 day of ageing at 80°C corresponds to approximately 10 years of natural ageing at 20°C. Knowing the time required for the fraction of broken bonds or folding endurance at 80°C to fall below a critical threshold, rendering the paper unusable, the time needed for the same effect to occur at 20°C can be calculated. In this way, the useful lifespan of a specific type of paper can be estimated. However, such conclusions should be regarded with caution, as experimental errors are significant. For a more comprehensive discussion on the limitations of the Arrhenius methodology presented here, interested readers are encouraged to consult the relevant literature [2]-[48]. Wilson and Parks [19] proposed using this methodology to classify different types of paper into permanence categories: less than 50 years, between 50 and 100 years, and several hundred years. Zou et al. [11] present plots of δ versus accelerated ageing time, illustrating the effects of temperature [11 p. 250, fig. 2] and relative humidity [11 p. 252, fig. 4] on the ageing rate. They also provide an Arrhenius plot on a logarithmic y-axis based on these results [11 p. 251, fig. 3].

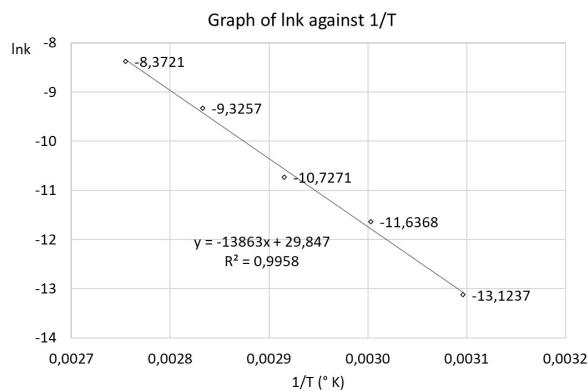


Fig. 8. Plot of the $\ln k$ against inverse absolute temperature ($1/T$)

V. APPLICATIONS

A. Effect of Acidity on the Ageing Rate of Paper - Deacidification

It was observed early on that while a significant portion of historical handmade papers, produced between the 15th and the middle of 19th century, retained good mechanical strength and acceptable color, most modern papers, manufactured from around the mid-19th to the late 20th century, deteriorated rapidly. As a result, many became unusable within just a few decades of their production.

The studies conducted by Barrow and Sproull [28] established the now widely accepted view that the primary cause of the degradation of modern papers was their increased acidity. This conclusion was supported by measuring the rate of loss in folding endurance among papers with varying acidity levels, using the methodology described earlier [28 p. 1083, fig. 7].

Taking this a step further, experiments using accelerated ageing were conducted to estimate the folding endurance loss rate in acidic versus neutralized samples of the same papers. The results showed that the acidic samples lost their strength rapidly, whereas the neutralized ones exhibited remarkable stability [28 p. 1083, fig. 7]. Additionally, it was found that the lower the pH (or the higher the acidity), the faster the hydrolysis of cellulose in the paper [11 p. 255, fig. 8]. Since Barrow's pioneering studies, this finding has been consistently confirmed. Acid hydrolysis of cellulose, the mechanism of glycosidic bond cleavage discussed earlier, is the fundamental chemical process responsible for the loss of mechanical strength in paper under acidic conditions.

Identifying the problem simultaneously provided its solution. *Deacidification*, by neutralizing the acidity of acidic papers, slows down the process of acid hydrolysis of cellulose, reducing the rate of mechanical strength loss and thereby extending the useful lifespan of acidic papers. Numerous deacidification methods have been proposed to achieve this goal, ranging from manual, time-consuming techniques used in small laboratories with minimal equipment [3]-[12]-[50-62] to automated mass deacidification processes requiring industrial-scale facilities [63-74].

The simplest and most widely used deacidification method, applied in most conservation laboratories worldwide, involves immersing paper in an aqueous solution of calcium hydroxide [3]-[62]. Calcium hydroxide is a base, reacting with and neutralizing the acids present in the paper.

The purpose of deacidification is twofold: to neutralize present acidity, and to deposit an alkaline buffer to guard against future acid accumulation. During the ageing process, the pH of paper gradually decreases due to the absorption of acidic pollutants from the atmosphere, the migration of acidic components from adjacent materials, and the

¹³ The E_a values reported in the literature typically range between 80–120 kJ/mol.

formation of organic acids as the paper deteriorates [2]-[41]. The excess deacidifying agent, known as the *alkaline reserve*, will neutralize any future acidity that may develop. However, deacidification cannot reverse the ageing process, it can only slow it down from the moment of treatment onward. Additionally, it offers long-term protection, making it a method of chemical stabilization. Nevertheless, it cannot restore the lost mechanical strength of the paper.

B. Permanent Paper

The expansion of the deacidification concept led to the realization that newly manufactured paper intended for long-term stability should be non-acidic and preferably, alkaline. This idea began to be gradually implemented in the paper industry from the 1980s onward.

Since the mid-18th century, with the industrialization of paper production, a new sizing system was introduced and replaced the traditional gelatin-based sizing system. This new system involved the use of potassium alum $[KAl(SO_4)_2 \cdot 12H_2O]$ or later aluminum sulfate $[Al_2(SO_4)_3]$ and a natural resin, rosin. Both aluminum sulfate and alum made paper acidic [8]-[75-78].

With growing awareness of the detrimental effects of high paper acidity, the industry began experimenting with new sizing systems that could function in alkaline conditions [79]. A major breakthrough came with the introduction of calcium carbonate as a filler material [4]-[8]-[80]. Calcium carbonate is a salt, but it has the important property of reacting with acids, forming calcium salts and carbon dioxide. This allows it to neutralize acids, functioning similarly to bases. Accelerated ageing tests demonstrated that calcium carbonate, in combination with modern non-acidic sizing systems, significantly enhances the chemical stability (*permanence*) of paper.

Furthermore, studies have shown that producing paper under alkaline conditions not only significantly extends its longevity but also reduces production costs, minimizes wear of manufacturing equipment, and results in a whiter final product [79]-[81]. Today, virtually all paper manufactured for writing and printing purposes is alkaline, containing calcium carbonate as a buffering agent. This paper typically has a pH between 7.5 and 10 and is classified as *permanent paper* due to its exceptional resistance to ageing, ensuring chemical and mechanical stability over extended periods [82]-[83].

The International Organization for Standardization (ISO) has established two key standards for paper longevity:

ISO 9706 [84] specifies the requirements for paper used in documents intended for permanent preservation. This type of paper must contain a minimum amount of calcium carbonate to ensure chemical and mechanical stability. The infinity symbol (∞) is used to designate paper that meets this standard.

ISO 11108 [85] applies to archival paper, meaning paper intended for frequent use and long-term preservation. This standard sets stricter requirements than ISO 9706, with a particular emphasis on the necessary mechanical strength to withstand handling over time.

C. pH of Materials Used for Long-Term Storage of Books and Archives

As discussed earlier, when cellulose is exposed to an acidic environment, its chains begin to break at random points through the process of acid hydrolysis [6]-[8]. This leads to a reduction in chain length, causing the gradual loss of paper strength over time [8]. The lower the pH, the faster the deterioration of paper strength. Experimental studies have also shown that during the ageing process, the pH of paper decreases [2]-[41] due to the generation of organic acids within the paper itself. Many of the byproducts of paper ageing are acidic gases or volatile compounds that behave like gases [86-89]. These substances can be absorbed by paper, meaning that direct contact between two sheets is not necessary for the transfer of acidic components. Simply coexisting in a closed or poorly ventilated space will eventually lead to the spread of acidity, accelerating the degradation process [90]-[91].

Accelerated ageing experiments conducted in sealed vessels have demonstrated that paper deteriorates significantly faster than in ovens with air renewal, confirming theoretical predictions. Furthermore, simply adding some alkaline paper to the ageing vessels resulted in a notable reduction in the ageing rate [27]-[89].

If acidic papers and cardboards, as well as glues and other materials, are used for the storage of archival materials and books, their volatile ageing byproducts will be transferred and absorbed by the original materials, accelerating their deterioration. However, even initially neutral papers tend to become acidic over time as they age [2]-[41]. Archival paper also undergoes progressive acidification during its lifespan. Therefore, if the papers and cardboards used for storage contain an *alkaline buffer*, there is potential for the neutralization of acids produced by the archival materials themselves. This alkaline buffer, typically calcium carbonate, offers dual protection by neutralizing acids produced during the ageing process of the stored material and shielding paper from acidic atmospheric pollutants [92].

Thus, the selection of appropriate storage materials not only provides passive mechanical protection but also actively contributes to the chemical preservation of archival materials, thereby significantly extending the lifespan of acidic and aged papers [93]. Conversely, acidic storage materials can rapidly degrade fragile archival documents, causing irreversible damage within a short period.

D. Evaluation of Paper Conservation Methods and Materials

Apart from deacidification, which aims at the chemical stabilization of paper and whose effectiveness was initially demonstrated using accelerated ageing methodology, there are numerous conservation interventions with various objectives. These include the chemical stabilization of both paper and a particularly corrosive type of ink (iron gall ink), disinfection, mending, the strengthening of mold-damaged and brittle paper, etc. [3].

Many of these methods are relatively new and have not yet stood the test of time. The same applies to many modern

synthetic materials. It is important to remember the destructive consequences of certain older conservation methods. In particular, the use of some synthetic resins (polymers) has caused irreversible damage, as their ageing behavior was not well understood at the time [94].

In current practice, before a new material or conservation method is adopted, its impact on the ageing rate of paper is thoroughly tested. Samples of treated paper (either subjected to the conservation method or treated with the new material under evaluation) undergo accelerated ageing tests alongside untreated control samples [1]-[17].

By comparing the ageing rates of treated and untreated samples, researchers can determine whether and how the tested method or material affects the degradation of paper. Such methods must be tested on multiple types of paper, including historical paper similar to the materials they are intended for. This approach has led to the rejection of several past conservation techniques and the adoption of improved methods and materials. Accelerated ageing can also be used for quality control and the optimization of conservation treatments in specific applications.

E. Effect of Temperature and Relative Humidity on the Ageing Rate of Paper - Storage Conditions

Chemistry demonstrates that an increase in temperature accelerates chemical reactions, a principle that also applies to the ageing of paper. Based on various published studies on artificial paper ageing [2]-[95]-[96] that followed the general methodology described earlier, Table IV was constructed. The relative ageing rates were calculated under the assumption that E_a equals 100 kJ. This value is representative of a paper grade exhibiting moderate stability. Using this assumption, the approximate relative ageing rates of this paper under different environmental conditions were estimated. The ageing rate at 20°C and 50% relative humidity (RH) was set to 1, and all other rates were calculated relative to this baseline.

Table IV: Relative ageing rates of a specific type of paper under various temperatures and relative humidities (50% and 75%). The calculation of these relative rates is based on the assumption that E_a equals 100 kJ, a value derived from a comparative study of multiple artificial ageing experiments on paper. [2]-[95-97].

T (°C)	0	5	10	15	20	25	30	35	40	45	50
RH: 50%	0.05	0.11	0.23	0.49	1.0	2.0	3.9	7.4	13.7	25.1	45.1
RH: 75%	0.08	0.16	0.34	0.74	1.5	3.0	5.8	11.1	20.6	37.7	67.7

From Table IV, it is evident that a simultaneous increase in temperature by 10°C and relative humidity by 25%—shifting from 20°C and 50% RH to 30°C and 75% RH—leads to a nearly sixfold increase in the ageing rate. For example, a type of paper with a useful lifespan of 600 years at 20°C and 50% RH would have its lifespan reduced to approximately 100 years under 30°C and 75% RH conditions. From the

perspective of natural ageing, maintaining lower temperatures and relative humidity levels significantly slows down the degradation process of paper.

Under conditions of low relative humidity and temperature (below 55-60% RH and 20-23°C), biological activity is also significantly reduced. Fungi, which pose the greatest threat, become inactive under these conditions, though they are not eradicated [98]. The activity of insects and rodents also declines. Controlling biological activity is another important reason for regulating the climatic conditions in storage areas [99].

However, relative humidity should not drop below 25%, as materials may become too dry, and if already in an advanced state of ageing, they may become brittle [100]. This sets a lower limit for relative humidity. Is there a lower limit for temperature as well? From a purely scientific perspective when considering the ageing rate, the answer is no. The minimum temperature for storage conditions is determined by many other factors, including economic, technical, and environmental [95].

Historic buildings, often used to house archives, libraries, and museums in Greece, other Mediterranean countries, and many regions worldwide, typically lack insulation and frequently exhibit structural defects. In other words, they are characterized by poor *building physics* [101]¹⁴. As suggested by Brokerhof et al. [101], “It is evident that it is impossible to control the climate inside buildings with a poor-quality building envelope, even with an advanced air treatment system. On the other hand, a building with high-quality building physics, barely requires climate control to create a stable climate inside. This is the starting point for sustainability.”

Achieving low temperatures depends on the quality of the building physics, the local microclimate, and the financial resources available for installing and operating a climate control system. In hot climates, maintaining very low temperatures is both challenging and costly [102]. Additionally, it depends on the location of storage areas. Underground, specially constructed well-insulated spaces free from moisture tend to remain cool even during summer months, making the maintenance of low temperatures in such environments relatively cost-effective. Naturally, it is essential to ensure these spaces are fully protected against the risk of flooding.

Successful efforts have been made to maintain acceptable climatic conditions with appropriate building design and minimal use of electricity [103]. By relaxing the requirement for highly stable conditions all year round, stipulated by older standards ($\pm 2^\circ\text{C}$ and $\pm 5\%$ RH), modern storage standards recommend a gradual shift in average temperature for energy conservation [104]-[105]. During winter, the average temperature can remain relatively low, while it can gradually increase as the warmer months

¹⁴ The term “building physics” refers to the construction quality, the presence or absence of insulation, the quality and materials used, as well as the impermeability of walls, windows, roofing, and other elements that passively influence the stability and independence of internal climatic

conditions from external ones. An example of poor building envelope quality could be a historic building lacking insulation, with single-glazed windows, and cracks or gaps in the masonry and window frames.

approach, up to a reasonable level suited to the local climate.

Modern standards, unlike older ones, do not prescribe fixed values with strict, minimal fluctuations in temperature and relative humidity [106]-[107]. Instead, they define acceptable ranges for these parameters and provide guidelines for selecting the most appropriate conditions for a specific cultural institution, considering among others financial, environmental, and climatic factors. For further details, interested readers are encouraged to consult the relevant literature and updated standards for archival, library, and museum storage environments [104]-[105]-[107-110].

VI. EPILOGUE

Accelerated ageing is a key tool for understanding the long-term implications of today's preservation decisions, choices that will significantly affect the future condition of our written cultural heritage. It provides a valuable means of assessing the effects of storage conditions and the relative merits of conservation treatments, particularly when long-term empirical data is unavailable.

The preceding discussion seeks to promote a clearer framework of communication between archivists, librarians, and conservators, with the aim of strengthening collaboration and enhancing the overall effectiveness of preservation strategies.

Beyond its technical objectives, this article is also intended to support understanding and dialogue across disciplines. It aims to make key scientific concepts, often found in the literature of conservation and preservation science, more accessible to professionals in archives and libraries. By presenting these ideas through simplified models and illustrative examples, it can serve as a useful reference for those involved in managing preservation strategies or evaluating related scientific publications.

In this way, the article contributes to interdisciplinary communication and training, helping bridge the gap between research and professional practice.

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