Rate of Paper Degradation
The Predictive Value of Artificial Aging Tests

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European Commission
on Preservation and Access, Amsterdam

2000

The European Commission on Preservation and Access gratefully acknowledges the financial support of the following institutions and organizations

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Preface

This publication is a translation of the Dutch report *Snelheid van papierverval/de betrouwbaarheid van prognoses op basis van kunstmatige-verouderingstests* (Porck 1999), resulting from a desk research project of the Koninklijke Bibliotheek (KB). The study was subsidized by the Ministry of Education, Culture and Science of The Netherlands, in the framework of *Metamorfoze*, the Dutch National Program for the Conservation of Library Materials. As a consequence of additional reviewing of the translated version, some corrections have been made in the original text.

In the preparation of this report, W.J.Th. Smit (Department of Conservation and Optical Technics, KB) offered valuable support. Useful contributions and comments were also made by H.J. Jansen and S. Pauk (Department of Research and Network Services, KB), and C.B.J. de Wolf (Collection & Research Division, KB).

Dr. D. Erhardt (Smithsonian Center for Materials Research and Education, Suitland, MD), Dr. H. Bansa (Bayerische Staatsbibliothek, München), Dr. J.C.M. van Eijndhoven (Rathenau Institute, The Hague), Dr. J. Down (Canadian Conservation Institute, Ottawa), Dr. V.D. Daniels (British Museum Conservation Department, London) and Dr. R.L. Feller (former coworker Carnegie Mellon Research Institute, Pittsburgh) are acknowledged for their contributions to the discussion and worthwhile suggestions.

Dr. J.B.G.A. Havermans (TNO Paper & Board Research, Delft), Dr. N.S. Baer (Fine Arts Institute, New York University), Dr. E. McCrady (Abbey Publications, Austin, TX) and Y. de Lusenet (ECPA, Amsterdam) critically reviewed the manuscript and offered many useful comments and recommendations.

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Summary

During artificial or accelerated aging a material is subjected to extreme conditions in a climate-chamber to try to speed up the natural aging process. Artificial aging tests are often used to determine the permanence of paper, i.e. its rate of degradation, as well as to predict the long-term effect of a conservation treatment. However, there are still many questions surrounding the actual predictive value of these tests. The present report, based on specialist literature and discussions with experts, aims at providing an overview of the current state of affairs concerning the possibilities and limitations of artificial aging analysis. The report is not only intended for conservation scientists involved in research in this field, but also for those responsible for the development of a conservation policy.

Since the fifties a great variety of artificial aging methods has been developed for paper, and the field of application of these methods in the practice of conservation of archival and library materials has broadened enormously. Nevertheless, fundamental and experimental research into the reliability of artificial aging analyses is only performed on a limited scale. An evaluation of this research indicates that the essential questions have not been resolved satisfactorily, and that findings have not resulted in the use of a generally accepted standard method. Differences in opinion still prevail, and there are evident disagreements with respect to the conditions under which artificial aging should preferably be performed.

In this report it is shown that the results of artificial aging tests currently available do not allow for a reliable determination of the rate of paper degradation in absolute terms. Meaningful application of the tests is limited to qualitative, relative statements on the expected effects of conservation treatments and on the role of the various reactions that contribute to the deterioration of paper.

The present report offers several practical recommendations and suggestions on alternative approaches to the question of how quickly paper deteriorates. It is also proposed to organize a meeting on this topic, for instance under the auspices of the ECPA or the CLIR. Such a meeting should help to break through the present deadlock characterized by uncertainty and doubt, and work out an agenda for a joint approach to the problem.

Introduction

Background

In conservation research methods are often being used that aim to accelerate the natural degradation of archival and library materials. In this way, scientists try to obtain more insight into the permanence of certain materials, the long-term effects of conservation treatments and the mechanisms of specific deterioration processes.

For these artificial or accelerated aging methods, a material is exposed in a climate-chamber to extreme conditions in terms of temperature and humidity for a certain period of time, during which the changes occurring in the material are measured. Subsequently, from the data obtained in this way, the rate at which the material in question will deteriorate under normal, natural conditions of storage is deduced. By comparison of the findings with untreated reference material, it is attempted to determine the expected effect of a certain conservation treatment. In addition, artificial aging tests can be used in order to study, or simulate, the physicochemical reactions of the natural aging process.

Questions

Although artificial aging tests have been used as a general research tool in the past decades, several serious questions with regard to the actual predictive value of these tests still exist. Both in the current specialist literature and in the more informal discussions in the conservation science community, the use of artificial aging, especially with regard to paper, is a subject of debate. The following are some of the questions that are being discussed:

1. Are the present (standard) methods for artificial aging, which in first instance have been developed for modern papers, suitable to study papers which have already undergone a natural and, depending on the storage conditions, also variable process of natural aging?
2. Are artificial aging tests meaningful for the determination of the actual rate of natural paper degradation, and thus suitable for making reliable predictions?
3. Can artificial aging analysis be used for the determination of the quantitative effect of a conservation treatment, or is the application of the results of this analysis limited to qualitative, relative interpretations?
4. To what extent are the reaction mechanisms of paper degradation which occur in artificial aging, equivalent to those under normal, natural storage conditions?

Objectives and set-up

The lack of consensus on the questions above presents a serious problem, both from a fundamental scientific point of view as for the practice of conservation. In order to motivate and plan conservation activities, and to reserve the necessary funds, it is important to have an accurate indication of the rate of paper decay, as well as a reliable insight into the medium- and long-term effect of a certain conservation treatment or measure.

For these reasons, the Koninklijke Bibliotheek has taken the initiative to address this issue, within the framework of the research activities of the national conservation program *Metamorfoze*. In planning the project, it was envisaged that the results should be useful for scientists directly involved in conservation research, as well as policy-makers in the field of preservation of archival and library materials.

The goal of the present report is to provide an up-to-date overview of the research and scientific debate on the theoretical basis and practical use of artificial aging. The final section of the report is devoted to a critical evaluation of the possibilities and limitations in the determination of the rate of paper degradation and other applications of artificial aging tests. Finally, several conclusions are drawn and some recommendations for the future formulated.

This study is also intended as a discussion paper for the committee on artificial aging of the working group 'Archive and Library Conservation', established by the Netherlands Institute for Cultural Heritage (NICH) in Amsterdam for the exchange and dissemination of knowledge and practical experience between conservators and conservation scientists.

Besides relevant specialist literature, the possibilities of the 'electronic highway' have also been explored in this project. By means of several issues of an 'aging newsletter', distributed by e-mail, it was tried to poll the opinions of a number of experts in the Netherlands and abroad. Both the literature and the discussion on Internet, which unfortunately only received limited response, clearly brought to light the current uncertainty and many points of disagreement.

Theory and practice

Principles and techniques of artificial aging

Just like all other organic materials in archives and libraries, paper is subjected to a number of fundamental deterioration processes. Under normal conditions of storage, these processes are very slow, but eventually and inevitably they still lead to well-known aging effects such as yellowing and loss of strength.

The most common reaction is the hydrolytic degradation of the cellulose molecules, the building blocks of paper, in which the presence of water (moisture) plays an essential role. According to standard reaction-kinetic principles, the rate of the hydrolytic process is determined by the temperature, the acidity (pH value), and the amount of moisture present in the paper. The moisture content is again dependent on the (relative) humidity in the storage environment.

The second deterioration process is the oxidative degradation of cellulose, primarily induced by the presence of oxygen in the environmental air. The third main mechanism of deterioration is the process of thermal degradation. This mechanism concerns the breakage of chemical bonds as a consequence of the temperature-dependent movements of the cellulose molecules and their constituent atoms.

Arrhenius-tests

The three deterioration reactions mentioned above take place simultaneously in the natural aging process of paper. Although the individual rates of these reactions vary, it is assumed that the overall rate of the degradation process is systematically related to the temperature, provided that the other environmental conditions are constant. Therefore, it appears possible to use the results of artificial aging at high temperatures to deduce the rate at which the deterioration process will take place at cooler, *casu quo* normal storage conditions.

The argument for the use of elevated temperatures in artificial aging relies on the fact that in general a reaction proceeds faster at higher temperatures, which makes it possible to observe its effects, in this case the loss of paper strength, more quickly than at room temperature. Such artificial aging experiments are sometimes called 'Arrhenius-tests'. In these tests, performed at different temperatures and a constant relative humidity of the environment (in a climate-chamber), a characteristic property of the paper, such as folding endurance or tear resistance, is measured at regular time intervals. By determining how many days of artificial aging (d) are needed at different aging temperatures (T) to obtain a certain measure of paper degradation (for instance a certain percentage of loss in folding endurance), a temperature-versus-'time needed' graphics can be made, i.e. an Arrhenius-plot. When the factor 'time' is plotted in a logarithmic scale (log d), and the temperature in a reciprocal scale (1/T), the Arrhenius-plot will theoretically show a straight line, reflecting a proportional relationship. By simply lengthening (extrapolating) this straight line to lower temperatures – that is below the temperatures at which the artificial aging was performed – it can be seen from the

Arrhenius-plot how much time would be needed for the same level of paper decay to occur at normal storage temperature. This procedure allows, in theory, the prediction of the rate of paper degradation at any temperature, thus also for instance at storage conditions below room temperature (cool storage).

Other uses
Except for the Arrhenius-test, the principle of the acceleration of chemical reactions is especially applied to predict the long-term effects of conservation treatments. In this case, the primary focus is not on the quantification of the influence of the treatment on paper permanence, but on the comparison of the effect of artificial aging between treated papers and untreated reference papers. On the basis of this comparison it is subsequently concluded whether or not the conservation treatment offers a significant benefit with regard to the aging resistance of the paper. For this purpose, instead of different high temperatures and measurements at regular intervals, fixed (standardized) aging conditions are used. The analyses of paper properties are often limited to one measurement before and one after the aging test. The examples of such tests presented in the next section will show that, in addition to the elevation of the temperature, other 'stimulating' conditions are applied in order to speed up the deterioration processes.

Finally, the acceleration of deterioration reactions is used to simulate certain types of damage in order to study the fundamental mechanism of the degradation. This approach enables the investigation of the relation between the visual appearance of the damage and the underlying deterioration reactions. In addition, simulating a specific type of damage under laboratory conditions offers a suitable alternative for the use of original collection materials in the development of conservation methods.

Methods and applications

The literature in the field of conservation research shows a great variety of artificial aging tests (Neimo 1964; Pravilova 1964; Rahn 1974; Brown 1991). In the first place, the objectives and as a consequence the ways of performance of these test strongly differ (Feller 1994a). Though not often indicated explicitly in the research reports, the three procedures mentioned above – Arrhenius-tests, comparative studies and simulation experiments – can be distinguished. Within these three groups a further subdivision is possible into different concrete applications.

In the second place, there are many differences in the actual methods themselves: the conditions under which the artificial aging is performed in the climate-chambers, which are sometimes constructed especially for these purposes. Among these methods are several international and national standards, such as:

- ISO-standards from the International Standards Organization (ISO 1982, 1985, 1986a,b);
- TAPPI-standards from the Technical Association of the Pulp & Paper Industry (TAPPI 1963, 1985a,b);

- ASTM-standards from the American Society for Testing and Materials (ASTM 1971, 1987);
- NEN-standards from the Netherlands Normalization Institute (NEN 1983);
- CSN- and AFNOR NFQ-standards from normalization institutes in respectively the Czech Republic and France (Hanus 1987, 1995, 1996; Daniel 1992).

In spite of the availability of these standards, other methods, often developed individually by research institutes, are used much more frequently in practice.

It should be pointed out here that the methods of artificial aging used in paper conservation are as a rule deduced from procedures developed in the paper industry for research on and production of special materials, such as heat- and fire-resistant types of paper and board (Back 1963, 1967a,b; Wink 1968; Fung 1969).

In the following table, a chronological overview is presented of a number of concrete and illustrative examples of artificial aging methods with their respective fields of application, selected from the specialist literature of the last decades. The descriptions list, in consecutive order, temperature (°C), relative humidity (RH in %), presence of other 'stimuli' (lighting, radiation, presence of gases such as oxygen [O_2], sulfur dioxide [SO_2], nitrogen oxides [NO_x], nitrogen dioxide [NO_2]), and if indicated, the duration of the aging treatment in days (d) or hours. If several aging methods have been used in combination, they are presented separated by a semicolon (;).

Additional notes: the notation 100/150 means: 100 or 150; 100-150 means: different values ranging from 100 to 150. If data on relative humidity (RH) are not given, it has to be taken into account that at high temperatures (>90 °C) climate chambers are comparable to ventilated ovens, and under these circumstances an RH value of 1-5% usually prevails.

method	application	reference
20°C, SO_2 treatment, 7d	influence of air pollutants	Langwell 1955
80°C, 70%RH, 5-30d	effect of paper strengthening with polymethyl acrylate	Yabrova 1958b
70°C, 50%RH, 10d; 100°C, 0/50%RH, 3d	permanence of different sorts of paper	Fink 1965
90°C, 0/50%RH, O_2 treatment, 1-24d	mechanism of aging, role of oxygen	Parks 1971a, 1971b,1972
105-150°C, 0%RH, O_2 treatment, <1d	mechanism of aging role of oxygen	Medved 1973
90°C,13d;35°C,lighting,12d; 90°C, radioactive radiation	effect (self)adhesive tapes	Skall 1974
100°C, 0/100%RH, O_2 treatment, 1-10d	role of oxygen and moisture in aging	Arney 1979
90°C, 50/100%RH, 7-35d	influence of washing with water	Tang 1979
60-120°C, 100%RH, O_2 treatment, 1-10d	role of oxygen and temperature in aging	Arney 1980
90°C, 100%RH, O_2 treatment and various pH-values,1-28d	role of oxygen and acidity [pH] in aging	Arney 1981
90°C, 50%RH, 7-21d	production test material, influence RH on folding endurance	Sclawy 1981
70-100°C, 2/50%RH, 3-179d	mechanism of aging	Mendenhall 1981
60-165°C; lighting and SO_2 treatment	mechanism of copper corrosion	Banik 1982
100°C, 7d	influence of cleaning with eraser powder	Pearlstein 1982
103°C, 3-24d	mechanism of aging	Hanus 1987
80°C, 10-81d, paper samples in sealed glass tubes	influence historical papermaking techniques	Barrett 1989
80°C, 65%RH, 12d	permanence of paper types	Ernst 1989
90°C, 40/60%RH (change in RH every 12 hours), 1-32d	influence of fluctuating storage conditions	Shahani 1989
90°C, lighting, 3-30d	effect cellulose ethers on discoloration	Feller 1990
90°C, 56%RH, 16d	effect cellulose ethers on folding endurance and acidity	Baker 1991
70°C, 0%RH, 35/42d; treatment with exhaust-fumes	influence of gelatin-sizing	Barrett 1992

Artificial Aging Methods and Applications (continued 1)

method	**application**	**reference**
80°C, 65%RH, 12d; 30°C,50%RH,lighting,ca.6d	influence of optical whiteners	Flieder 1992; Leclerc 1993a
90°C,60%RH,7d;28°C,80%RH, SO_2 and NO_2 treatment,7d	effect mass deacidification, effect air pollution on deacidified paper	Daniel 1992
105°C, 3d	influence of storage in inert gas (argon)	Havermans 1992
90°C, 50%RH, 28d	production of test material, effect conservation of transparent-paper	Hofmann 1992; Reyden 1992
80°C, 60%RH, 7-28d; gamma radiation	simulation of degradation, production of paper for conservation purposes	Inaba 1992
90°C, 55%RH, 40d	production of test material for non-destructive analysis	Priest 1992
90°C, 50%RH, 20d	influence of bleaching by means of moisture and light	Schaeffer 1992
80°C,95%RH,1-32d;22°C,65% RH, SO_2 and NO_x, 1-28d	effect mass deacidification, effect air pollution on deacidified paper	Banik 1993
50°C, 65%RH, 10d; 30°C, 50%RH,lighting,ca.12d	permanence of photocopies	Leclerc 1993b
90°C, 65%RH, 30d; 30°C, 50%RH,lighting,ca.12d	stability of printing inks	Maraval 1993
90/50°C,80/35%RH(change in T,RH every 6 hours), 6d	effect of encapsulation in polyester film	Pauk 1993
60/100°C, 1-100d	effect of washing with water on gum arabic as binding agent	Daniels 1993
50-90°C, 30-38%RH, 2-1372d	mechanism of discoloration, role of temperature and RH	Vitale 1993
80°C, 65%RH, 6-24d	NEN-2728 and DIN-6738 standards for permanent paper	Berkhout 1994
90°C,50%RH,12d;23°C, 50%RH, SO_2 and NO_2, 4d	effect mass deacidification, effect air pollution on deacidified paper	Havermans 1994,1995
105°C, 3-24d; 80°C, 65%RH, 3-24d	effect of disinfection with ethylene oxide and gamma radiation	Hofenk 1994
80°C, 50%RH, 14-99d	production test material, effect mass deacidification	Kaminska 1994
90°C, 50%RH, 12d	effect mass deacidification with Battelle and DEZ system	Pauk 1994
103°C, 24d	influence of storage in boxes	Hanus 1995,1996
90°C, 50%RH, 12d	effect mass deacidification with Bookkeeper system	Pauk 1996a

Artificial Aging Methods and Applications (continued 2)

method	application	reference
90°C, 50%RH, ca. 21d	effect of various strengthening methods	Bansa 1997
80°C,65%RH, 6-24d	influence of paper composition and production technique	Letnar 1997
90°C, 35/80%RH (change in RH every 3 hours)	simulation ink corrosion, effect of treatment methods	Neevel 1997

Natural and artificial aging

The application of artificial aging is assumed to be justified. However, this hypothesis must be verified in practice. This verification offers in addition the possibility to make adjustments to the artificial aging test used. In this way the relevance of the test in predicting the natural aging process can be optimized. On the basis of the literature and discussion with experts, an overview of the developments and current state of affairs will be presented below.

At the end of the nineteenth century the possibility to intensify the natural deterioration of paper artificially by dry heat treatment was already described (Herzberg 1899). In 1926 the Swedish Government Testing Institute compared the permanence of different papers after exposure to elevated temperatures and sunlight (Hall 1926). At the end of the twenties, research of the National Bureau of Standards (USA) showed that dry air at 100 °C resulted in the strongest and best reproducible aging effects. This finding formed the basis of the artificial aging method of 72 hours at 100 °C (Rasch 1929) which was often used since then. Later empirical studies, for instance by Wilson et al. (1955) and Barrow (1963), suggested that this artificial aging corresponds with 18-25 years of natural aging at room temperature. The applicability of a dry, thermal artificial aging test for the prediction of paper permanence was also confirmed in other studies dating from the fifties and sixties (Royen 1958; Barrow 1964).

Only a few investigations have been performed into other possibilities to stimulate natural aging (Kantrowitz 1940; Launer 1943). This may indicate that the heat treatment was generally accepted as a valuable method. In this context it is interesting to note that in the current research program of the American Society for Testing and Materials (ASTM) the focus will be on 'alternative' means in the development of artificial aging tests, aiming at the use of lighting and air pollutants (Arnold 1996).

In the course of the fifties it became increasingly clear that the tests so far performed by means of dry heat do not take into account the fact that aging under laboratory conditions fundamentally differs from the natural aging process taking place at normal relative humidities. For this reason the suspicion grew that these tests could well lead to the wrong conclusions (Richter 1956; Yabrova 1958a; Graminski 1970,1979; Wilson 1971; Arney 1979). In addition, gradually more doubts arose on the usefulness of artificial aging performed at only one raised value of the temperature. It was acknowledged that the reactions responsible for paper deterioration need not all be related to temperature in the same way, partly because of differences in activation energy, i.e. the minimal energy required for a reaction to take place (Stamm 1956; Royen 1957; Browning 1968,1969; Gray 1969,1977; Smith 1970; Graminski 1979; Arney 1980; Bansa 1984).

In first instance, these questions led to the conviction that an approach according to the Arrhenius-principles -artificial aging at one, more realistic value of relative humidity and at different temperatures, and extrapolation of the findings to room temperature- would be more justified from a scientific point of view (Baer 1974; Lindström 1989). However, there is still disagreement about the most suitable values for temperature and relative humidity. As a consequence, the variation in the conditions used for artificial aging is enormous. In a literature overview from 1964 the following ranges are given: temperatures between 22 and 160 °C, relative humidities from 1% to 100%, and a duration of treatment between one hour and 180 days (Neimo 1964).

Problems

Nowadays the issue of temperature and relative humidity in artificial aging is still topical. It is worth mentioning that the well-known 'rule of thumb' that a rise in temperature of 10 °C results in a doubling of the reaction rate is considered not to apply to the deterioration process of paper (Erhardt 1989). In addition, arguments have been put forward that the absolute quantity of moisture in the air rather than the relative humidity is the factor determining the rate of deterioration (Bansa 1998b; Down 1998).

Moreover, there is another fundamental problem: while the field of application of the Arrhenius-principles concerns the reaction-kinetics of chemical transformations, the complex properties of paper which are often registered in artificial aging, such as folding endurance, tear resistance and paper discoloration, cannot actually be simply and unambiguously related to the chemical composition of the paper in question. Nonetheless, there are indications that under certain conditions the rate at which such characteristics change still exhibits a systematical relation to the chemical processes which are taking place during artificial aging. Therefore in practice, in the analysis of artificial aging, it is assumed that the principles of chemical reaction-kinetics do indeed apply (Luner 1969; Lindström 1989; Zou 1996; Erhardt 1998).

Complexity of the material

That there are still a number of snags in the application of artificial aging tests is also attributed in the literature to the complexity of the material itself. In the case of paper, we are not only dealing with cellulose containing vegetable fibers, but often with other compounds as well, such as lignin, fillers, pigments and sizing agents. As a consequence, paper should be considered as a multi-component system rather than as a singular substance, which complicates interpretation of the results of an artificial aging test (Baer 1977; Down 1998).

Another complicating factor concerns the way in which the paper is exposed to the aging conditions. Research findings from the Library of Congress (Shahani 1989,1991,1993,1994), the Netherlands Institute for Cultural Heritage (Roelofs 1994), the Koninklijke Bibliotheek (Pauk 1996b) and the Australian National Library (Brandis 1997) have clearly indicated that paper in stacks, c.q. books, shows a different aging behavior compared to single, loose sheets. Some of these studies have shown that both under artificial and natural aging conditions, deterioration is more severe at the center of a stack of paper than at the regions more to the outside. In addition, damage surveys performed in several library collections had already demonstrated that in areas with relatively low air pollution, paper in book blocks can be significantly more acid (showing a lower pH value) in the middle of the pages than in the margins (Smith 1972; Porck 1990). This also points to a stronger aging process in the center of paper stacks than in the outer parts. This 'stack-versus-single sheet' phenomenon in the aging of paper is considered of major importance and will be studied in more detail in the future (Down 1998).

Comparison of artificial and natural aging

Only few investigations have been performed in which artificial and natural paper aging are directly compared (Wilson 1955,1980; Royen 1957, 1958). In 1928 the National Bureau of Standards tested various chemical and physicomechanical properties of different writing papers by means of artificial aging (100 °C, 3 days), while a part of the original material was stored in a box for future research. From the results of later studies it was concluded that only a qualitative relation between the effects of artificial aging and the natural aging of the original paper that had taken place in the meantime could be demonstrated (Wilson 1955).

In a more recent study Wilson and Parks (1980) compared the changes in properties after 36 years of natural aging with the results of artificial aging (100 °C, 3 days) of the original material in 1937. With some types of paper, changes in folding endurance, tear resistance and several chemical characteristics that had occurred in natural aging appeared to correspond reasonably well with those obtained by means of artificial aging. Other types of paper showed more deviations.

An elegant approach of the comparison between natural and artificial aging can be found in the current research of Erhardt and other co-workers from the Smithsonian Center for Materials Research and Education (SCMRE) in Suitland, USA (Erhardt 1987,1988,1993,1995). The starting-point in their studies is that the results of artificial aging can only lead to a reliable predictions with regard to natural aging if the artificial aging method speeds up the rate of deterioration of paper without fundamentally changing the nature of the process. This means that every individual reaction involved in the decay ought to be accelerated by the same factor, keeping the ratio between the reaction velocities constant. Extended comparison of the effects of different artificial aging methods, monitoring the various degradation reactions separately by means of sensitive measuring equipment, led to the interesting conclusion that the nature of the paper deterioration process is a function of relative humidity (RH) rather than temperature. By changing the temperature at a constant RH value, the rate of the deterioration process is altered without fundamental changes in the process itself, so that the same level of aging can be obtained at different temperatures, simply by adjusting the duration of the treatment. However, a change in RH, leaving the temperature constant, results in a different proportion between the rates of the individual reactions and consequent changes in the distribution of reaction products, which makes it impossible to reach the same level of aging at different RHs. If one extends this conclusion, drawn from investigations on different artificial aging methods, to the simulation of natural aging, it would seem advisable to apply the artificial aging at the same relative humidity as at the storage conditions under which the natural aging takes place.

Comparison of identical copies of books

Another interesting line of research in this context is the comparison of identical copies of books which, as part of separate library collections, have been stored under different conditions and as a consequence show different stages of deterioration. Although this kind of research does not result directly into recommendations for artificial aging, as it actually concerns a comparison of two different ways of natural aging, relevant information can still be obtained. Besides offering more insight into environmental effects on the rate of natural aging of paper, these studies results also indicate which environmental factors are responsible for the difference in aging condition and thus in rate of decay.

In this way, comparative investigations of this kind can indirectly be of importance for the development of reliable artificial aging methods. The study on pairs of books from the New York Library (NYPL) and the Koninklijke Bibliotheek (KB), performed by the KB in collaboration with TNO Paper & Board Research (Delft), showed that the paper in the NYPL copies deteriorated faster due to a higher concentration of the air pollutant sulfur dioxide, in combination with low and/or fluctuating relative humidities in the NYPL storage rooms (Pauk 1996b; Havermans 1997a). Indications for a role of air pollution in the development of differences in book pairs have also been found in previous comparative studies (Hudson 1967; Smith 1972; Baer 1986).

Long-term storage projects

Useful information on the rate of paper deterioration and on the storage conditions which play an important role in this respect, can also be expected from a current research project of the General State Archives in The Hague (Feber 1998). In this study, which the Koninklijke Bibliotheek has joined through the SALTO-project ('*s*tudie *a*angaande *l*ange *t*ermijn *o*pslag', study concerning long-term storage), identical archive and library materials and other test papers are stored at two locations, one of which is provided with an installation to filter air pollutants. By means of continuous registration of environmental conditions like temperature, humidity and concentrations of air pollutants, as well as frequent analysis of the quality of the stored material in both storage rooms, interesting data will become available in the course of time. These data will offer more insight into the long-term effects of air filtering, and they will also give a realistic indication of the rate at which the various types of paper deteriorate under different degrees of air pollution.

Damage surveys

The analysis of the data resulting from damage surveys has resulted in other comparative studies. In the last decades these surveys have been performed worldwide in many archives and libraries, primarily in order to gain a better understanding of the nature and extent of paper degradation. By repeating such surveys after a certain time interval, re-examining the same test materials, and by comparison of the results with the findings of the earlier tests, an indication can be obtained of the rate with which certain paper properties have changed in the meantime. For example, a damage survey from 1990 at the Koninklijke Bibliotheek was repeated on a limited scale in 1996, clearly showing that a measurable decrease in folding endurance had occurred already in the course of six years. However, due to the limitations of this investigation, definite statements about the rate of paper decay could not be made (Hol 1990; Pauk 1997).

Historical approach

Finally it is worth mentioning that an historical approach also appears to create possibilities for acquiring insight into the natural aging rate of paper. In recent studies into historical sources on the inferior quality of nineteenth-century Dutch paper, this approach has been elaborated. It is argued that the results of contemporary mid-nineteenth century scientific investigations into paper quality and comparison of these data with the findings of present-day examination of the same material, traced in archival collections, in principle produce useful indications of the rate of paper decay (Grijn 1996, 1998; Porck 1996).

Discussion

Evaluation

'Artificial aging' is a test in general use and the term a common one in conservation research. This seems to suggest (i) that there exists a normal, natural aging process, (ii) that this process can be artificially accelerated without changing in its nature, and (iii) that by using artificial aging tests meaningful information on this natural aging process can be obtained. The present study shows that all of these suggestions can be seriously questioned.

Insufficient knowledge on natural aging
In the first place there is no generally valid description of the normal, natural aging process of paper. There are so many paper-dependent internal factors, partly as yet unknown, as well as external factors varying with time and place that influence the stability of paper that it seems impossible to predict unambiguously the natural aging of a particular sheet of paper (Roberson 1981; Wilson 1983; Fellers 1989; Feller 1994a,b; Gurnagul 1994; Norris 1994). Still more complex is the situation in a stack of paper, like a book or a file, let alone a complete library or archive collection. A reliable judgement on the nature and rate of natural aging can actually be made only afterwards, i.e. deduced in retrospect. Moreover this is only possible if detailed information is available on the original material condition of the object and on the storage conditions to which the object has been exposed in the course of time. The examples of comparative studies on identical copies of books stored under different conditions, mentioned in the preceding section, show that such a reconstruction of the storage history of collections is no sinecure.

Simulation by stimulation?
In the second place it is probably only wishful thinking that natural aging of paper can be simply accelerated by exposure to artificial, laboratory conditions, and that in this way certain forms of damage, familiar from the conservation practice, can be reliably simulated. The complex character of the natural aging process, which is partly still obscure, can never be completely achieved experimentally. Even if natural aging could be simplified to the effects of the paper degradation reactions as known today, it is highly doubtful that artificial stimulation of these reactions, by means of certain adjustments of environmental parameters in a climate-chamber, will result in an identical condition of the paper in the short term as would result from the natural aging process in the long term. Such an assumption is as yet insufficiently verified by scientific research. The current uncertainty in this regard is clearly illustrated by the multitude of standard and 'home' methods for artificial aging mentioned above. Artificial aging is performed at different temperatures, different humidities, sometimes in the presence of air pollutants or other 'stimuli' and sometimes not, and for different periods of time. Even the standard methods for artificial aging, developed in order to counteract this large variation, do not resolve this: the international ISO-standards, for example, developed in the eighties, leave the possibility open to age artificially at different temperatures and relative humidities.

Relevance of artificial aging

All this implies that quite a few objections can be put forward against the idea that artificial aging presents relevant data for the determination of the nature and rate of the natural aging of paper. The studies into the reliability, i.e. predictive value of artificial aging, as described above, have not yet resulted in a conclusion that is generally endorsed, nor into the use of a generally accepted method. The findings of the few investigations performed in this respect have not yet sufficiently proven in practice that the theoretical basis for the application of artificial aging is indeed valid. Nor have these studies resulted in adjustments of current methods. The limited scale of the research into the comparison of artificial and natural paper aging is remarkable. However complex this kind of investigation may be, it is one of the few possibilities to attune the procedure of artificial aging to the natural aging process. In the field of leather conservation, for example, this approach appears to have been rather successful (Larsen 1994).

Problem of reliability

The small number of research projects that have tried to verify the predictive value of artificial aging analysis strongly contrasts with the widespread use of this analysis in practice. Even so, this does not mean there is no concern about the issue. The problem of the reliability of artificial aging has been raised frequently in the specialist literature (Arney 1980; Bansa 1984, 1992, 1998a; Mokken 1987; Erhardt 1988; Ströfer-Hua 1990; Wypych 1990; Brown 1991; Feller 1994a; Shahani 1994; Havermans 1997b). The present insecurity about the value of artificial aging can for instance be illustrated from the fact that the international standard ISO-9706 for permanent paper, in which a prediction of the durability and permanence would be extremely desirable, does not include an artificial aging test (Berkhout 1994).

In view of the different objectives with which artificial aging is applied, the Arrhenius-test, aimed at a quantitative statement on permanence, i.e. rate of paper decay, will probably be the most debatable. In the case of other applications, like the determination of the qualitative effect of a conservation treatment, the investigation of a certain degradation reaction or the simulation of specific kinds of damage, the strict prerequisite for the Arrhenius-test, namely that all natural deterioration reactions must be accelerated by one and the same factor, does not need to be fulfilled. In these cases it will be more likely that artificial aging may produce trustworthy indications. Still, here too reservations should be made because on the basis of the present state of knowledge, it cannot be excluded that unrealistic effects will occur as a consequence of undesirable reactions, i.e. reactions absent in the natural aging process.

Deadlock

With regard to artificial aging there seems to have developed a kind of deadlock in the conservation science community, and sometimes even an obvious reluctance to exchange experience and to strive for a joint approach to tackle the problem. This reluctance, which perhaps also caused the limited response to the discussion planned for the present study, can be explained to some extent. A critical discussion of such crucial and historically deeply rooted issues may lead to changes in opinions that can have far-reaching consequences. It is possible that decisions taken in the past, either in conservation policy or conservation practice, based on the results of artificial aging tests, are undermined in such a discussion. On the other hand the current deadlock is a cause for serious regret, as it obstructs a solution of the many questions on artificial aging which are still left unanswered. In the meantime the natural aging of paper proceeds without mercy.

Conclusions

In summary, four conclusions can be drawn on the basis of this evaluation of the current state of affairs with respect to the theory and practice of artificial aging of paper:

The rate of paper deterioration and other quantitative aspects of the natural aging of paper, such as durability and permanence, cannot be reliably predicted by means of the present artificial aging tests.

The application of artificial aging can be relevant and meaningful with respect to:
a. the determination of the qualitative effect of a certain paper conservation treatment;
b. fundamental research into paper degradation reactions;
c. the simulation of specific types of paper damage.

The amount of research into the predictive value of artificial aging of paper is not sufficient and the results are not adequately used for the improvement of artificial aging methods.

Reluctance in discussion and maintenance of a *status quo* characterize the conservation science community with regard to the problem of artificial aging. As a consequence the solution of this problem is seriously hindered.

Recommendations

The present study is intended as an overview and evaluation of the current state-of-the-art rather than an elaboration of solutions. However, several recommendations can be made which may contribute to the improvement of the current situation of uncertainty and doubt.

The exchange of knowledge and experience in the field of artificial aging analysis of paper among conservation scientists and policy makers needs to be improved. A special workshop on this subject, under the auspices of the European Commission on Preservation and Access, the Koninklijke Bibliotheek in cooperation with the Netherlands Institute for Cultural Heritage, or the Council on Library and Information Resources may offer a suitable framework for such a discussion.

Research to verify the hypothesis that artificial aging produces meaningful data to predict the effects of natural aging of paper should be continued and subsequently be ‘translated’ in improved, i.e. more reliable artificial aging tests.

Comparative investigation of natural and artificial aging of paper can be performed in parallel with long-term studies into natural paper deterioration, by means of artificial aging treatments and subsequent analysis of part of the initial test material. The findings can be compared in a later stage, by a next generation of conservation scientists, with those of the naturally aged material.

It is recommended that statements on the rate of the natural aging of a certain kind of paper are not only limited to predictions for the future by means of artificial aging. In some cases, a retrospective approach is possible, using historical sources that present detailed information on the quality of a specific type of paper in the past.

In the application of the current artificial aging tests for the determination of the relative effects of a certain paper conservation treatment, for the investigation of a specific degradation mechanism, or for the simulation of a certain form of paper decay, a 'mild' artificial aging method is to be preferred because of the inevitable risk of undesirable degradation reactions, that is to say reactions that do not occur under natural conditions. Artificial aging conditions should therefore in principle be kept as closely as possible to normal, natural conditions.

As false expectations may be evoked by the term 'artificial aging' it should be considered:

a. to limit the use of the term 'aging' to the genuine process of natural aging;
b. to use another term for the so-called artificial aging test, like 'artificially induced degradation (AID-) test', which reflects the way in which these tests are commonly used in practice: choosing and adjusting the environmental conditions in such a way that a measurable deterioration of the material takes place in a relatively short time.

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