NBS TECHNICAL NOTE 1118

U.S. DEPARTMENT OF COMMERCE / National Bureau of Standards

Stone Consolidating Materials— A Status Report

NATIONAL BUREAU OF STANDARDS

The National Bureau of Standards[1] was established by an act of Congress on March 3, 1901. The Bureau's overall goal is to strengthen and advance the Nation's science and technology and facilitate their effective application for public benefit. To this end, the Bureau conducts research and provides: (1) a basis for the Nation's physical measurement system, (2) scientific and technological services for industry and government, (3) a technical basis for equity in trade, and (4) technical services to promote public safety. The Bureau's technical work is performed by the National Measurement Laboratory, the National Engineering Laboratory, and the Institute for Computer Sciences and Technology.

THE NATIONAL MEASUREMENT LABORATORY provides the national system of physical and chemical and materials measurement; coordinates the system with measurement systems of other nations and furnishes essential services leading to accurate and uniform physical and chemical measurement throughout the Nation's scientific community, industry, and commerce; conducts materials research leading to improved methods of measurement, standards, and data on the properties of materials needed by industry, commerce, educational institutions, and Government; provides advisory and research services to other Government agencies; develops, produces, and distributes Standard Reference Materials; and provides calibration services. The Laboratory consists of the following centers:

> Absolute Physical Quantities[2] — Radiation Research — Thermodynamics and Molecular Science — Analytical Chemistry — Materials Science.

THE NATIONAL ENGINEERING LABORATORY provides technology and technical services to the public and private sectors to address national needs and to solve national problems; conducts research in engineering and applied science in support of these efforts; builds and maintains competence in the necessary disciplines required to carry out this research and technical service; develops engineering data and measurement capabilities; provides engineering measurement traceability services; develops test methods and proposes engineering standards and code changes; develops and proposes new engineering practices; and develops and improves mechanisms to transfer results of its research to the ultimate user. The Laboratory consists of the following centers:

> Applied Mathematics — Electronics and Electrical Engineering[2] — Mechanical Engineering and Process Technology[2] — Building Technology — Fire Research — Consumer Product Technology — Field Methods.

THE INSTITUTE FOR COMPUTER SCIENCES AND TECHNOLOGY conducts research and provides scientific and technical services to aid Federal agencies in the selection, acquisition, application, and use of computer technology to improve effectiveness and economy in Government operations in accordance with Public Law 89-306 (40 U.S.C. 759), relevant Executive Orders, and other directives; carries out this mission by managing the Federal Information Processing Standards Program, developing Federal ADP standards guidelines, and managing Federal participation in ADP voluntary standardization activities; provides scientific and technological advisory services and assistance to Federal agencies; and provides the technical foundation for computer-related policies of the Federal Government. The Institute consists of the following centers:

> Programming Science and Technology — Computer Systems Engineering.

[1] Headquarters and Laboratories at Gaithersburg, MD, unless otherwise noted; mailing address Washington, DC 20234.

[2] Some divisions within the center are located at Boulder, CO 80303.

Stone Consolidating Materials—A Status Report

James. R. Clifton

Center for Building Technology
National Engineering Laboratory
National Bureau of Standards
Washington, D.C. 20234

Sponsored by:

National Park Service
U.S. Department of Interior
Washington, D.C. 20240

t. Technical note

U.S. DEPARTMENT OF COMMERCE, Philip M. Klutznick, Secretary
Luther H. Hodges, Jr., Deputy Secretary
Jordan J. Baruch, Assistant Secretary for Productivity, Technology, and Innovation

U.S. NATIONAL BUREAU OF STANDARDS, Ernest Ambler, Director

Issued May 1980

National Bureau of Standards Technical Note 1118
Nat. Bur. Stand. (U.S.), Tech. Note 1118, 52 pages (May 1980)
CODEN: NBTNAE

U.S. GOVERNMENT PRINTING OFFICE
WASHINGTON: 1980

For sale by the Superintendent of Documents, U.S. Government Printing Office, Washington, D.C. 20402
Price $3.50
(Add 25 percent additional for other than U.S. mailing)

ABSTRACT

Information on types of stone consolidating materials, their performances, and uses are critically reviewed. Processes responsible for the deterioration of stone and criteria for selecting stone consolidants are also reviewed. The main function of stone consolidants is to reestablish the cohesion between particles of deteriorated stone. In addition, a good consolidant should meet performance requirements concerning durability, depth of penetration, effect on stone porosity, effect on moisture transfer, compatibility with stone, and effect on appearance.

Stone consolidants can be divided into four main groups, according to their chemistry. These groups are inorganic materials, alkoxysilanes, synthetic organic polymers, and waxes. Epoxies, acrylics, and alkoxysilanes are currently the most commonly used consolidating materials. Certain waxes have been found to have excellent consolidating abilities. Waxes, however, tend to soften and to accumulate grime and dust. Inorganic materials and some organic polymers have a tendency to form shallow, brittle layers near the surfaces of stone. These treated layers often separate from the untreated stone. Alkoxysilanes have excellent penetrabilities and are considered by some stone conservators as the most promising consolidating materials. However, a universal consolidant does not exist and many factors must be considered in selecting a consolidant for a specific stone structure.

Key Words: Conservation; consolidating materials; deterioration of stone; preservation; stone; stone consolidation.

TABLE OF CONTENTS

Page

TABLE OF CONTENTS (Continued)

1. INTRODUCTION

This report is a critical literature review of the types, performances, and uses of stone consolidants. Processes responsible for the deterioration of stone and criteria for selecting stone consolidants are also reviewed. Stone consolidants are generally considered [1-4] to be deeply penetrating materials which have the ability to re-establish the cohesion between particles of deteriorated stone. A variety of materials have been used in attempting to consolidate deteriorated stone, including inorganic chemicals, organic polymers, alkoxysilanes, resins, and waxes, either by themselves or in combinations. Because of an increasing worldwide interest in preserving historic structures and an apparent acceleration in the rate of stone decay, the use of stone consolidants is growing [5,6]. However, concern is growing over the use of unproven materials on important historic structures. This review has been carried out under the auspices of the National Park Service to assess the current status of stone consolidation technology.

Stone preservatives whose chief functions are to prevent the ingress of moisture into stone, i.e., water repellents, have been covered elsewhere [7-9] and are not included in the present report. Treatments with water repellents often do not have long term preservation effects and may even accelerate stone decay through two major processes [10,11]. First, water will often collect behind the treated stone and upon evaporation of the water any salts in solution will be deposited and crystallize in the untreated stone. This may lead to spalling of the treated stone from the untreated stone. In the second process, because of differences in thermal expansive properties of the treated and untreated portions of the stone, shear stresses may be generated that eventually result in interfacial delamination. Sleater [12] recently tested over 50 stone preservatives[1] and found that none of them gave satisfactory performance.

Numerous reports have been published on stone preservation and consolidation and an extensive bibliography covering the literature through 1963 was prepared by Lewin [13]. Excellent sources of information on the mineralogy, weathering, and conservation of building stones are the books by Winkler [14], Schaffer [15], and Warnes [16]. Because of the growing interest in the preservation of historic stone structures, several international symposia have been held recently on the deterioration and conservation of stone [17-20].

In this report the deterioration and consolidation of limestone and sandstone mainly will be addressed. This is because most of the problems encountered in the preservation of historic stone structures located in the United States are associated with these types of stone. Problems are also being encountered with the preservation of marble in countries where it has been extensively used, such as Italy and Greece.

2. DETERIORATION OF BUILDING STONE

The processes leading to the deterioration of building stone have been the subject of numerous publications [11, 14-16, 21-23]. Therefore, stone deterioration is only briefly reviewed herein for the purpose of providing a basis for understanding the performances of stone consolidants. The factoı considered to be among the leading causes of building stone deterioration

---

[1] In recent times the term "stone preservatives" has been used to identify water repellent and/or shallow penetrating coatings.

include salt crystallization, aqueous dissolution, frost damage, microbiological growth, human contact, and original construction. In this review, however, only a few cases were found in which the cause of stone deterioration was unequivocally determined. All too often a speculative approach was used in the analysis of stone deterioration, in place of a more scientific diagnostic method.

2.1 Salt Crystallization

Crystallization of salts within the pores of stones can generate sufficient stresses to cause the cracking of stone, often into powder fragments. This process is considered to be the major cause of stone deterioration in many parts of Western Europe [24-26]. Closely related to the crystallization of salt is damage caused by salt hydration and by differential thermal expansion of salts [14]. The resistance of stone to salt damage is dependent on the pore size distribution and decreases as the proportion of fine pores increases [11]. Crystallization damage caused by highly soluble salts, such as sodium chloride and sodium sulfate, is usually manifested by powdering and crumbling of the stone's surface [11]. Less soluble salts such as calcium sulfate form glassy, adherent films which cause spalling of a stone's surface [16].

A major source of salts in urban environments is the reaction between air pollutants and stone. For example, limestone can react with sulfur dioxide to ultimately produce calcium sulfate. Other sources of salts include ground water [27], airborne salts [27], sea spray [28], chemical cleaners [29], and deicing salts [30].

2.2 Aqueous Dissolution

Carbonate sedimentary stones (e.g., limestone and dolostone), carbonate-cemented sandstone, and marbles are types of stone that are susceptible to dissolution by water acidified with dissolved carbon dioxide, sulfur dioxide, and nitrogen oxides [31]. It has been reported [32, 51] that the rainwaters in many urban areas in the United States and Europe are sufficiently acidic to accelerate the weathering of exposed building stone. In areas where the rainwater is relatively free from pollutants, the dissolution of most common building stones is usually not a serious problem [30].

2.3 Frost Damage

Certain stones which are exposed to freezing temperatures and wet conditions may undergo frost damage. The frost susceptibility of a stone is largely controlled by its porosity and pore size distribution [33,34]. Of stones with a given porosity, those with the smallest mean pore size will generally be the most susceptible to frost damage. Frost resistance also generally decreases with increased available porosity [15], i.e., pore volume which is accessible to water. The frost resistance of a stone is often assessed from its saturation coefficient,[2] with stones having saturation coefficients less than 0.8 being generally immune to frost damage [35].

Some European stone conservators [11,26] believe that in their countries frost damage is not an important process in the deterioration of stone. They regard frost damage as a secondary process, e.g., frost damage may be responsibile for the final fragmentation of stone damaged by other processes, such as salt crystallization. However, because of the use of possibly more frost-susceptible stone and more severe climates,

---

[2] Saturation coefficient is defined as the ratio between the natural capacity of a stone to absorb water and its absolute porosity [35].

frost damage may be an important factor in the northern part of the United States [36,37].

2.4 Microbiological Growth

The attack of stone by a variety of plants and animals has been reported [14] including roots of plants, ivy vines, microorganisms, boring animals, and birds. Of these, microorganisms appear to be the most destructive. Some types of bacteria, fungi, algae, and lichens produce acids and other chemicals which can attack carbonate and silicate minerals [14, 38]. It appears that under certain environmental conditions attack by microorganisms can be a serious problem [39-41]. However, it seems that many conservators feel that such instances are uncommon and that microorganism growth usually takes place in stone which had been partially deteriorated by other processes.

2.5 Human Contact

Because of an increasing interest by the public in historic structures, the effects of human contact upon the condition of stone, as well as all other building materials, is of growing concern. For example, stone floors are gradually worn by foot traffic, stones are damaged by people either collecting souvenirs or poking into soft stone [28], and graffiti removal has become an important maintenance problem [42-48]. It is conceivable that human contact may become a major problem challenging the ingenuity of both stone conservators and maintenance specialists.

2.6 Original Construction

The durability of stone structures also depends on factors encountered during their original construction, including proper design, good construction practices, and proper selection of materials. Unfortunately, these are factors over which the preservation scientist has no control. However, the same mistakes should not be repeated in repairing or restoring historic structures. For example, normal steel and cast iron anchors, dowels, reinforcing rods, etc., were often used in the construction or repair of stone structures. Certain ferrous metals are susceptible to corrosion which can lead to the cracking and spalling of stonework [11]. Therefore, non-corroding material should be selected, e.g., epoxy-coated steel [44], certain types of stainless steel [45], or non-corroding non-ferrous alloys [11].

A large portion of stone durability problems are the consequence of using poor quality stone in the original construction. Riederer has suggested [46, 47] that air pollution is often blamed for stone deterioration in Germany which actually should be attributed to the natural weathering of poor quality stone. The use of poor quality sandstone in completing the Cathedral of Cologne [48,49] is presenting acute conservation problems. In another example, a poor quality dolomite limestone was used in the construction of the British Houses of Parliament [50] which eventually had to be replaced with a more durable limestone [15]. It is doubtful that a stone consolidant can make a poor quality stone durable and, as with the above example, stone replacement can be at times the most rational approach [52].

3. PERFORMANCE REQUIREMENTS FOR CONSOLIDANTS

Performance requirements for stone consolidants have gradually emerged as understanding of the factors controlling their performance has improved. These requirements can be divided into two major categories. The first category, which we will term primary requirements, consist of invariable performance requirements that consolidants must fulfill regardless of the specific application. In the remaining category are secondary performance requirements. These are requirements imposed on a consolidant by a specific application. The distinction between the two categories will become clearer as they are discussed.

3.1 Primary Performance Requirements

Primary performance requirements for stone consolidants are applicable to essentially all stone consolidants regardless of the specific use. These requirements are based on the premise that the main functions of such materials are to restore the cohesion, physical properties, and appearance of a deteriorated stone to near its original condition. Considerations are given in the requirements to the necessary levels of performance of consolidants for consolidation, durability of the consolidated stone, depth of penetration, effect on stone porosity and moisture transfer, compatability of consolidant with stone, and effect on appearance. These performance considerations could become the basis for stone consolidant specifications.

3.1.1 Consolidating Value

The most important function of a stone consolidant is to re-establish cohesion of the particles of deteriorated stone [1-4]. Methods which have been used to obtain a qualitative assessment of the consolidating value of stone consolidants include the measurement of the tensile strength of treated unweathered stone [25, 53], measurement of surface hardness of treated weathered stone [54,55], and the abrasion resistance of treated unweathered stone [56]. A more quantitative test needs to be developed, which incorporates the testing of standard deteriorated stone. A standard deteriorated stone may merely be finely crushed stone, which is then treated with consolidants and its tensile strength measured. The tensile strength of the treated crushed stone could be compared to the strength of the original stone.

Only one recommendation was found in this review for what constitutes an acceptable performance level for consolidation value. Gauri has recommended [54] that the compressive strength of a treated weathered stone should be at least 10 percent above that of the untreated and unweathered stone. However, increasing the strength of weathered stone substantially above that of unweathered stone may accelerate the decay of the unweathered stone [68] , unless the complete structure is treated with the consolidant.

3.1.2 Durability of Consolidated Stone

The consolidated stone should generally be as durable as the unweathered stone. If the consolidated stone is substantially less durable than unweathered stone, it may be advantageous to replace the deteriorated stone with new stone. In addition, treated stone should weather (in terms of appearance) at nearly the same rate as the original stone, to retain, as closely as possible, the authentic appearance of the stone. Torraca [1] has suggested that it is not justified to demand that a consolidant should protect a stone forever from any environment.

Durability of a consolidated stone is dependent on several factors, including the durability of the consolidant, compatibility of the consolidant with the weathered stone, properties of the consolidated stone, and the environment. The complex interaction among these factors makes the development of appropriate accelerated durability tests difficult. A methodology is given in ASTM E-632 [69] which can be useful in the design and development of rational accelerated durability tests for consolidants.

3.1.3 Depth of Penetration

Past experience with stone consolidants has shown that their ability to penetrate weathered stone is one of the main factors controlling their performance [3, 16, 25, 57-60]. Superficially-penetrating consolidants tend to fill the pores of stone surface layers, thereby reducing the permeability. This may result in the accumulation of moisture and salts behind the treated layers [58, 61]. Furthermore, interfacial delamination often occurs because of a marked difference in the thermal properties of the treated and untreated stone [10, 11, 58]. Price has suggested [58] that a good consolidant should be able to penetrate a weathered porous stone to a depth of at least 25 mm. This should result in a gradual transition in the thermal and mechanical properties from the exterior treated surface to the inner layer of untreated stone. A slightly different requirement has been given by Torraca [1]. He proposed that a stone consolidant should penetrate a weathered stone to a depth that all incoherent material is solidified and attached to the sound core of the stone.

Properties of a stone consolidant which affects its ability to penetrate a specific stone at a given temperature include its viscosity [57, 59, 61], surface tension [59], the rate at which gel or precipitate is formed [16], method and conditions of application [2, 63], and rate of evaporation of any solvent [66]. Tammes and Vos [67] developed the following relation for the horizontal transport of liquid through porous materials;

$$X = {\frac{\sigma r}{2N}}^{1/2} t^{1/2} = At^{1/2}$$

where X is the displacement of the liquid front, σ is the surface tension of the solution, r is the average capillary radius, N is the viscosity of the liquid, and t is time. A is a measure of the permeability of the porous material to a particular liquid. Munnikendam [59] showed that this equation can be used to estimate the penetration ability of a consolidant into a specific stone.

This survey revealed that a need exists for the development of a standard test to measure the penetration of consolidants into stone. This standard test should specify the testing temperature and relative humidity, condition and size of the stone specimen, methods for applying and curing consolidants, and techniques for measuring the depth of penetration.

3.1.4 Stone Porosity

As previously mentioned, the porosity and pore size distribution of a stone can have a major effect on its durability. For example, the resistance of a given type of stone to frost damage and to salt damage decreases as the proportion of fine pores increase [11, 33]. Therefore, a stone consolidant which reduces the size of large pores but does not close them may be harmful. For example, Dukes [2] found that silicone ester consolidants decreased the frost resistance of portland stone used as gravestones by increasing the number of small pores.

A method which can be used to determine the effects of a stone consolidant on pore size distribution was described by Clifton et al. [70]. In their work, the pore size distributions of both impregnated and unimpregnated hardened cement pastes were measured using a mercury porosimeter. Both pressurization and depressurization studies were performed. Pressurization results give an indication of the total open porosity and pore size distribution. Information on the shape and continuity of pores is obtained by depressurization work. Pressurization studies on stone have been performed by Biscontin and Pavan [71] and by Alessandrini et al. [72].

3.1.5 Moisture Transfer

Many stone preservatives and stone consolidants have performed poorly because they form a surface film which impedes liquid water migration through the treated stone, but allows water vapor to pass. This can lead to a situation where water evaporates behind the treated stone leaving deposits of salts [59, 61]. Because water vapor can pass in and out of the stone, the deposited salts may rehydrate and possibly be converted into different crystal forms. Further, larger crystals may grow through the dissolution of small crystals and reprecipitation on larger crystals. These processes may result in the disruption of the microstructure of the stone and breakdown of cohesion between stone particles [14]. Further, impeding the passage of water vapor may increase the susceptibility of a stone to frost damage and to thermal shock.

If a material produces a film which prevents the passage of both water and water vapor, large amounts of moisture could accumulate in a structure. In addition to decreasing the resistance of the stone to frost damage, excess moisture in a structure could cause the rotting of wood, corrosion of metals, and degradation of plaster and roofing materials.

Munnikendam has suggested [73] that a consolidant should be hydrophilic to allow moisture to pass through the treated stone. In addition, he suggested that the water vapor transmission of treated stone should not be decreased by more than 30 percent compared to untreated stone.

The water absorption and water vapor permeability of stone, untreated and treated, can be measured by following the procedures of ASTM C97 [74] and ASTM C355 [75], respectively.

3.1.6 Compatibility of Consolidant with Stone

Experiences with stone consolidants have demonstrated [16, 61] that they should be compatible with stone to form a durable composite. Specific compatibility requirements should include the following:

1. Cured consolidants should have thermal-dimensional properties similar to those of sound stone [16, 73]. Otherwise, delamination of the consolidated stone from the untreated stone could occur [76], especially if the stone is subjected to thermal shock. In addition, the cured consolidant should not become brittle [16, 76, 77].

2. The consolidant should not severely disrupt the microstructure of the stone [16]. For example, if the crystals formed from the precipitation of an inorganic consolidant exhibit crystal growth sufficient tensile stresses may be produced to cause the development of microcracks, and ultimately macrocracks in the matrix. Further, Marsh [50] pointed out that replacing a constituent of stone with another of a larger molecular volume can cause dilation stresses leading to cracking.

   The effects of a few inorganic consolidants, e.g., hydrofluoric acid, depend on their reacting with the constituents of the stone to form insoluble products. Many conservators [5, 16, 76, 78] are opposed to the use of such materials because the reaction products usually fill the voids and pores, thereby sealing the surfaces.

3. Consolidants should not form by-products which can be harmful to the stone. Many of the inorganic consolidants are precipitated as a result of the reaction between two dissolved salts [1, 16, 50]. In addition to the precipitate, at least one soluble salt is formed, much of which is deposited in the stone as the water evaporates. These soluble salts can damage the stone through

recrystallization processes and/or produce unsightly effloresence on the treated stone's surface.

3.1.7 Effect on Appearance

Ideally the application of a stone consolidant would not cause any change in the appearance of a stone. Most inorganic consolidants, however, produce a white deposit within the voids and pores of a stone, the color of which may not match the color of the stone. While organic consolidants usually form transparent polymers when cured, they can change the reflective properties of a stone [79]. Further, the optical properties of organic polymers may gradually change because of their degradation by photochemical processes, oxidation by oxygen and ozone, and attack by air pollutants.

The permissible extent of change in appearance accompanying the use of a consolidant is probably best dealt with by the conservator responsible for the preservation of a structure. This is because the extent of the change in appearance is partially controlled by the interactions between a consolidant and a specific stone, and also by the environment. Furthermore, the location of the deteriorated stone within a structure, the extent of stone deterioration, the nature of the change in appearance, and the importance of a structure should be considered in deciding what constitutes a permissible change in appearance.

In laboratory evaluations of the performance of stone consolidants, the change in appearance of a standard stone specimen should be quantitatively determined. The preparation of test specimens, exposure condition and evaluation methods should be standardized. Then the results of such a standard test could form the basis for selecting promising materials for specific applications. Gauri et al. [54] developed a laboratory performance test for organic consolidants in which the absorbancy of a polymer at 254 mu was compared to that of bisphenol-A epoxy resin.

Several methods can be used in the laboratory and in the field to measure the change in appearance of stone caused by consolidating materials. In a laboratory evaluation of preservatives, Sleater [12] instrumentally compared the color and gloss of treated and untreated stone specimens following the methods given in ASTM D 2244 [80] and ASTM D 523 [81]. He also recommended that a visual estimation of color differences be made. The Munsell color system [82] can be used in both the laboratory and field to determine color changes. Winkler [83] has developed a promising method to measure rapidly the reflectance of stone using a photographic light meter and reflex camera.

3.2 Secondary Performance Requirements

Secondary performance requirements are those requirements which may be imposed in addition to the primary performance requirements because of specific preservation problems encountered at certain structures. For example, the leading cause of stone deterioration in England is salt crystallization [24-25]. Therefore, in England the capacity of stone consolidants to encapsulate salts or to otherwise mitigate the effects of salts is an important consideration in their selection. Arnold and Price [84] have observed that certain consolidants facilitate the extraction of salts from stone, which could be more important than their ability to immobilize salts.

Other attributes which may be required of stone consolidants in certain situations include the ability to prevent further microbiological growth, to greatly increase the resistance of stone to abrasion by foot traffic, and to rebond large stone fragments.

4. STONE CONSOLIDANTS

In this review, stone consolidating materials are divided into four main groups, according to their chemistry. These groups are inorganic materials, alkoxysilanes, synthetic organic polymers, and waxes. Considerations of their performance are based on the requirements described in Section 3.

4.1 Inorganic Materials

Inorganic stone consolidants were extensively used during the 19th century and still are occasionally being used. Most inorganic consolidants produce a white insoluble phase within the voids and pores of a stone, either by precipitation of a salt or by chemical reactions with the stone. It has been rationalized that the development of a new phase similar in composition to the matrix of a stone will bind together the particles of deteriorated stone. For example, consolidants which result in the formation of a silica phase should be used to consolidate sandstone, and calcium carbonate or barium carbonate used to consolidate calcareous stones such as limestone. In practice, however, little concern is given to chemical compatibility between the consolidants and stone.

Little success has been achieved in consolidating stone with inorganic materials, and in some cases their use has greatly accelerated stone decay [16, 50, 78]. Some of the reasons given for the poor performance of inorganic consolidants are their tendencies to produce shallow and hard crusts [16, 60, 76], the formation of soluble salts as reaction by-products [1, 16, 60, 86, 87], growth of precipitated crystals [50], and the questionable ability of some of them to bind stone particles together [6, 85]. Of these, the most difficult problem to overcome is the formation of shallow hard surface layers by inorganic consolidants because of their poor penetration abilities. Precipitation processes are often so rapid that precipitates are formed before the inorganic chemicals can appreciably penetrate the stone. A method, referred to as precipitation from homogeneous solutions, has been developed to obtain deeper penetration of stone by some inorganic consolidants. This method is discussed in Section 4.1.2.

4.1.1 Siliceous Consolidants

Siliceous consolidants are materials which have been used to consolidate sandstone and limestone through the formation of silica or insoluble silicates.

4.1.1.1 Alkali Silicates

Both nonstoichiometric dispersions of silica in sodium hydroxide and soluble alkali silicates have been used to conserve and consolidate stone. When dispersions of silica in sodium hydroxide solutions are applied to a stone, silica is deposited [16, 88]. If sodium hydroxide is not removed by washing, it can react with carbon dioxide or sulfur trioxide to form sodium carbonate or sodium sulfate, respectively. These salts can cause unsightly effloresence and salt crystallization damage. In addition, it seems that sodium hydroxide can react with the constituent of some stones, thereby accelerating stone deterioration [16].

Silica can be precipitated by the reaction between sodium silicate, as well as postassium silicate, and acids such as hydrochloric acid and arsenic acid [16, 88, 89]. However, these reactions result in the formation of soluble salts such as sodium chloride and sodium arsenate. If the sodium silicate-arsenic acid mixture is used to consolidate limestone, crystalline calcium arsenate can be produced by a reaction between calcium carbonate and arsenic acid. The crystalline calcium arsenate appears to damage limestone by anistropic crystal growth [16].

Insoluble silicates have been precipitated in stone by alternate treatments of sodium silicate and a variety of salts such as calcium chloride [16, 85, 88, 91] and zinc carbonate [90]. Colloidal silicates are first produced which eventually become crystalline [16], while soluble salts are produced as by-products. Impervious surface layers are also produced which trap water beneath [92]. Apparently, the silicates precipitate relatively rapidly so that they are deposited near the surfaces of the treated stones.

Even with all the problems associated with the use of alkali silicates, they are still occasionally being applied [5]. Recently, the successful use of soluble silicates was reported [93]. However, the overwhelming evidence clearly indicates that alkali silicates should not be used for stone consolidating purposes.

4.1.1.2 Silicofluorides

Both hydrofluosilicic acid and soluble silicofluorides have been used to preserve and consolidate stone. Hydrofluorosilic acid should not be used on limestone as it reacts vigorously with calcium carbonate to form crystalline calcium silicofluoride, carbonic acid, and carbonate salts [91]. The reaction occurs upon contact of the acid with the limestone producing a shallow crust with little consolidating value. Hydrofluorosilic acid reacts more slowly with siliceous-based sandstones to form a cementitious material, but again only the surface is hardened. Hydrofluorosilic acid has a tendency to discolor both limestones and sandstones, especially if they contain iron [16].

Many soluble types of silicofluorides, such as magnesium, zinc, and aluminum, have been applied to limestone. Resulting products are silica, insoluble fluoride salts and carbon dioxide, which are formed near the surface of the limestone. Therefore, only the surface is hardened, which eventually exfoliates [52, 76, 94]. Soluble silicofluorides also react with calcareous sandstones and again only a hardened surface is obtained. Further, soluble salts are formed when both limestone and calcerous sandstone are treated with silicofluorides [60]. These soluble salts have caused damage through salt recrystallization processes [86]. Penkala [95] recently carried out a systematic study of several stone treatments and also found that fluorosilicates were not effective consolidators.

4.1.2 Alkaline Earth Hydroxides

4.1.2.1 Calcium Hydroxide

Aqueous solutions of calcium hydroxide (its saturated solution is often called limewater) have been used for many centuries to protect and consolidate limestone [96]. Calcium hydroxide itself does not appear to consolidate stone, but when in solution or a wet state it reacts with atmospheric carbon dioxide to form insoluble calcium carbonate, which may bind particles of calcareous stones together. The solubility of calcium hydroxide is only about 1 gram per liter at room temperature [97], therefore repeated applications are necessary to produce sufficient calcium carbonate to consolidate stone. Furthermore, unless very dilute solutions are used, only the calcium hydroxide deposited near the surface of a stone is carbonated. This happens if the dense calcium carbonate being formed at the surface fills the pores and voids in the stone. This severely impedes the migration of carbon dioxide through the treated surface to the interior of the stone.

The newly produced calcium carbonate is susceptible to the same deterioration processes as the calcareous stone. For example, it can react with sulfur trioxide to form calcium sulfate, which is relatively soluble compared to calcium carbonate. Therefore, the treated stone may not be protected against further weathering. However, the treated stone may eventually gain the authentic appearance of the weathering stone.

Conflicting opinions have been given of the effectiveness of the calcium hydroxide process. Some conservators [8, 16] have felt that while treatment with calcium hydroxide causes no harm, little permanent consolidation is obtained, while others [50, 96, 98, 99] have recommended the use of lime water to protect limestone from weathering and to consolidate them. The effectiveness of freshly prepared slaked lime (calcium oxide mixed with water) in consolidating statues at the Wells Cathedral in England is being investigated by Professor Baker [100]. He is applying 38 mm thick layers of slaked lime to statues, which are being removed several weeks later. Some consolidation appears to be occurring.

Apparently, repeated limewater and slaked lime treatment can gradually consolidate limestone, but such processes are only economically feasible for small objects.

4.1.2.2 Strontium and Barium Hydroxides

Similar to calcium hydroxide, strontium and barium hydroxides will react with carbon dioxide to form insoluble carbonates. Again, only the hydroxide near surface of a stone is usually carbonated. However, unlike calcium sulfate, strontium and barium sulfates are insoluble and thus the application of strontium and barium hydroxides may reduce the weathering of stone exposed to polluted environments.

The early work on the use of barium hydroxide to conserve stone was performed by Church [101-103]. Initially, excellent results were obtained. However, only a surface hardening was being obtained and eventually the barium carbonate or barium sulfate layer exfoliated [16, 50, 76, 92]. The exfoliation problem has been attributed not only to the formation of a dense impervious surface layer, but also to anisotropic crystal growth of barium carbonate and barium sulfate [16, 50].

Lewin [104] and Sayre [105] have developed methods intended to precipitate barium carbonate and barium sulfate deeply within a stone. These methods are based on a process known as precipitation from homogeneous solution [106]. In this process the material to be precipitated and the precipitating chemicals are present in the same solution. For example, barium carbonate is precipitated from an aqueous solution of barium hydroxide and urea [104, 108]. The urea slowly undergoes hydrolysis producing ammonia and carbon dioxide. The liberated ammonia and carbon dioxide dissolves in the water forming ammonium carbonate which raises the pH of the solution. When a certain pH is reached, barium hydroxide reacts with the carbonate ion and barium carbonate is precipitated. The reaction rate can be controlled so the barium carbonate precipitate forms days after a stone is treated. The slow formation of barium carbonate is reported to give a crystalline solid solution with the calcite crystals of calcareous stone. Barium sulfate can be precipitated in a stone by an analogous method. An aqueous solution of a barium monoester of sulfuric acid hydrolyzes slowly when a base is added, releasing barium and sulfate ions [106].

The precipitation of barium carbonate and barium sulfate by homogeneous solution precipitation methods is clearly a promising approach. To date, however, only experimental testing has been carried out and little is known concerning the long-term consolidating effectiveness of this approach. Warnes [16] and Marsh [50] have both suggested that crystalline inorganic precipitates, such as barium carbonate and sulfate, do not have long term consolidating value. Also the precipitates of barium carbonate and barium sulfate have a larger molecular volume than calcite and appear to exhibit anisotropic crystal growth [16, 50]. It should not be assumed that deteriorated stone will have sufficient empty volume to accommodate these precipitates. Therefore, until more is known of the long-term effects of barium carbonate and barium sulfate on the durability of stone, they should be regarded as experimental materials which should not be applied to important historic structures.

4.1.3 Other Inorganic Consolidants

Many other inorganic materials have been used in attempts to conserve and consolidate stone, including zinc and aluminum stearates [16, 28, 50, 95], aluminum sulfate [16, 50, 106], phosphoric acid [50], phosphates [50], and hydrofluoric acid [5]. Hydrofluoric acid appears to have a consolidating effect because it removes deteriorated stone, thereby leaving a sound surface. A saturated aqueous solution of calcium sulfate has been recently used to consolidate a stone consisting of a conglomerate of microfossils cemented by gypsum [107].

4.2 Alkoxysilanes

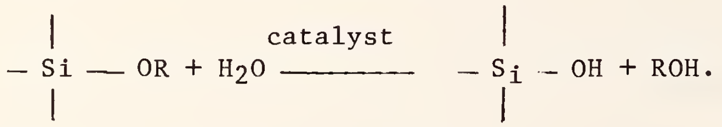
4.2.1 Uses and Developments

Alkoxysilanes are regarded by many stone conservators [8, 25, 59, 99, 109-114] as being among the most promising stone consolidating materials for siliceous sandstones. The feasibility of using alkoxysilanes to consolidate calcareous stone is also being studied [115-116]. The main reasons that alkoxysilanes are being highly regarded are their abilities to penetrate deeply into porous stone and the fact that their polymerization can be delayed until deep penetration has been achieved [25, 58, 99, 109, 110, 112]. In addition, they polymerize to produce materials similar to the binder in siliceous sandstone.

The use of alkoxysilanes for consolidating stone is not a recent development. For example, Laurie [117] received a patent in 1925 for producing such a material to be used for stone consolidation. Other early researchers on the use of alkoxysilanes to consolidate stone are Cogan and Setterstrom [118-119]. Alkoxysilanes have been commonly used since around 1960 in Germany [5]. Recently, a promising alkoxysilane consolidating material was developed at the UK Building Research Establishment, called "Brethane" [112].

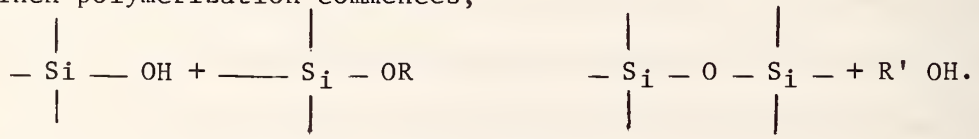
4.2.2 Alkoxysilane Chemistry

Alkoxysilanes are a family of monomeric molecules which react with water to form either silica or an alkylpolysiloxane. Three alkoxysilanes are commonly used to consolidate stone. They are tetraethoxysilane, triethoxymethylsilane and trimethoxymethylsilane [109]. Tetraethoxysilane is an example of a silicic acid ester [110]. Their polymerization is initiated by a hydrolysis reaction,



$$-\overset{|}{\underset{|}{\mathrm{Si}}}-\mathrm{OR}+\mathrm{H_2O}\xrightarrow{\text{catalyst}}-\overset{|}{\underset{|}{\mathrm{Si}}}-\mathrm{OH}+\mathrm{ROH}. \qquad (2)$$

Then polymerization commences,



$$-\overset{|}{\underset{|}{\mathrm{Si}}}-\mathrm{OH}+-\overset{|}{\underset{|}{\mathrm{Si}}}-\mathrm{OR}\qquad-\overset{|}{\underset{|}{\mathrm{Si}}}-\mathrm{O}-\overset{|}{\underset{|}{\mathrm{Si}}}-+\mathrm{R'OH}. \qquad (3)$$

where R = CH_3 (methyl), C_2H_5 (ethyl)

and R' = H, CH_3, C_2H_5

Polymerization continues until all the alkoxy groups have been liberated and either an alkylpolysiloxane or silica is produced. Silica is produced by the polymerization of a silicic acid ester. An alkylpolysiloxane is formed by the polymerization of other types of alkoxysilanes. An acidic catalyst, e.g., hydrochloric acid, is used to increase the rate of hydrolysis (equation 2). The alkoxysilanes are diluted with solvents to reduce their viscosities. Thus, their reaction rate and depth of penetration into stone can be controlled. It is claimed that their consolidating ability can be increased by using a mixture of alkoxysilanes [110].

Some confusion appears in the literature regarding silicon esters, silicones and alkoxysilanes. Silicon esters are partially polymerized alkoxysilanes which still have ester groups attached to silicon. Silicones are polymerized alkoxysilanes which are dissolved in organic solvents, and used as water repellents [110].

4.2.3 Performances of Alkoxysilanes

Price [58] and Weber [111] have observed that alkoxysilanes can penetrate porous stones to a depth of between 20 to 25 mm. The newly developed Brethane has been reported [112] to penetrate as deeply as 50 mm. No noticeable polymerization occurs with Brethane for at least 3 hours after it is mixed with a solvent and catalysis [58, 109]. The large sizes of unpolymerized alkoxysilane molecules, no doubt, will prevent them from entering the smaller pores of a stone.

Marschner reported [120] that alkoxysilanes improved the resistance of sandstone to sodium sulfate crystallization. However, she also observed that their performance varied from sandstone to sandstone and also depended on the compatibility between the solvent and the specific stone being treated. Similar findings were reported by Moncrieff [115] who studied the consolidation of marble. Snethlage and Klemm [121] observed in a scanning electron microscope analysis of impregnated sandstone that a polymerized alkoxysilane appeared to fill the space between sandstone grains and form a continuous coating. However, polymerized alkoxysilanes are reported [25, 58,110, 115] to have little effect on moisture passage in stone and the frost resistance of stone. Some slight changes in the color of treated stone have been observed [122, 123]. For example, statutes on the Wells Cathedral have become more dull grey following treatment with an alkoxysilane [123]. Further, a treated stone panel on the Cathedral has acquired a slightly more orange tone than adjacent untreated panels.

Once a section of stone is treated with alkoxysilane, it will probably weather differently than the untreated stone. Thus, unless most of the visible parts of a structure are similarly treated, the contrast between the treated and untreated stone could become very noticeable.

Strength improvements of around 20 percent have been reported [25, 110] when sandstone specimens were impregnated with alkoxysilanes. The ability of alkoxysilanes to consolidate deteriorated stone in the field, however, has not been unequivocally demonstrated. Further, it appears that the performance of alkoxysilanes varies from stone to stone.

Even if alkoxysilanes are found to be effective consolidants, their high cost [28, 112] will probably limit their use to statues and smaller-sized stone objects.

4.3 Synthetic Organic Polymer Systems

Two general types of synthetic organic polymer systems are used to consolidate stone. In the first, monomeric organic molecules are first polymerized, dissolved in appropriate solvents, and then applied to stone. They are deposited within the voids and pores of the stone as the solvent evaporates. The second type are monomeric organic molecules, either pure or dissolved in a solvent, which are polymerized within the voids and pores of a stone. Viscous monomers are diluted with solvents so that deep penetration can be achieved [57]. Solvents which evaporate rapidly (most common organic solvents), however, have been found to draw organic consolidants back to the surface of a stone, resulting in the formation of impervious hard surface crusts [57, 61]. Munnikendam [59] has recommended that organic consolidants should be selected whose solidification does not depend on solvent evaporation.

Both thermoplastics and thermosets have been used to consolidate stone. A thermoplastic is a material which can be reformed by the application of heat without significant changes in properties. Examples of thermoplastics are poly(vinylchloride), poly(ethylene), nylon, poly(styrene) and poly-(methylmethacrylate). A thermoset is a material which is formed into a permanent shape by the application of heat, and once formed, cannot be remelted or reformed. Polyester, epoxy, and polyurethane are examples of thermosets. Methylmethacrylate can be converted into a thermoset by copolymerization with a three dimensional cross-linking material.

The use of synthetic organic polymer systems to consolidate stone is a recent development, dating back to around the early 1960's. Therefore, little is known regarding the long-term performance of these materials. Some organic consolidants have been found to improve significantly the mechanical properties of deteriorated stone. Many organic polymers are susceptible to degradation by oxygen and ultraviolet radiation, but this would only affect the materials on the surface of a treated stone [1]. Riederer reported [5] that the surfaces of some stone structures in Germany which had been consolidated with organic polymers in 1965 had exhibited deep channel erosion by 1975. Apparently, water gradually eroded the consolidated surface and once the surface was pierced, erosion proceeded rapidly into the untreated stone.

4.3.1 Acrylic Polymers

Methylmethacrylate and to a lesser extent butylmethacrylate have been used to consolidate concrete [64, 65] and stone [58]. These monomers can be applied solvent-free to porous solids and can be polymerized in situ. An excellent source for information on their polymerization as well as on polymer-impregnated concrete is the report by Kukacha et al [64]. Methylmethacrylate has been polymerized into poly(methylmethacrylate) by heating with an initiator, by gamma radiation, and at ambient temperature by combination of promoters and initiators [64, 124]. For thermal polymerization, the chemical initiator (catalyst) azobis(isobutyronitrile) has been found to be effective [125]. Heating blankets could be used to thermally polymerize methylmethacrylate or other monomerics applied to a stone structure. Polymerization by radiation is only feasible if carried out in special chambers because of the radiation hazards. Chemical promoters convert initiators into free radicals at ambient temperatures. Then these free radicals induce the polymerization of methylmethacrylate. Munnikendam [61] used N,N-dimethyl-P-toluidine to decompose benzoyl peroxide into free radicals. He found, however, that oxygen inhibited the subsequent polymerization reaction of methylmethacrylate. Better success probably could be achieved by using azobis(isobutyronitrile) as the initiator [125].

Where deep or complete impregnation and complete polymerization was achieved, methylmethacrylate and other acrylates have been shown to improve substantially the mechanical properties and durability of porous materials such as concrete [64]. However, incomplete impregnation, with acrylates may result in the formation of a distinct interface between treated and untreated stone [120].

Polymer-impregnated concretes based on acrylics are classified as brittle materials based on their stress-strain curves [64, 125, 126]. Stone consolidated with methylmethacrylate and other acrylics can be expected to exhibit a similar brittle behavior.

Methylmethacrylate, no doubt, can harden the surface of a stone and effectively consolidate the stone if both deep penetration and complete polymerization are achieved. Similar to the case with alkoxysilanes, however, stone impregnated with methylmethacrylate will probably weather differently than untreated stone. In addition, erosion through the treated stone [5] could contribute to the development of an unsightly appearance.

4.3.2 Acrylic Copolymers

Copolymers are produced by the joining of two or more different monomers in a polymer chain [127]. A commercially available acrylic copolymer used for stone consolidation is copolymerized from ethylmethacrylate and methylacrylate [38, 121]. Other acrylic copolymers which have been studied for stone conservation include copolymers between acrylics and fluorocarbons [128, 129] and between acrylics and silicon esters [59, 121].

The acrylic copolymers are dissolved in organic solvents then applied to stone. As discussed earlier, unless very dilute solutions are applied to a stone solvent evaporation will tend to draw the acrylic copolymers back to the surface. Then, even if diluted to the lowest concentration that will give some consolidation, their solutions still may have viscosities which impede their penetration into stone.

4.3.3 Vinyl Polymers

Several vinyl polymers have been studied or used for conservation and consolidating of stone including poly(vinylchloride) [54, 130], chlorinated-poly(vinylchloride) [130], and poly(vinylacetate) [38, 54, 130, 131]. These polymers are dissolved in organic solvents and then applied to stone. Photochemical processes could release chlorine from the chloride polymers, which could damage stone [130]. Poly(vinylacetate) has been found to produce a glossy stone surface [130]. If not carefully applied and if not sufficiently diluted, use of the vinyl polymers undoubtedly will result in the formation of impervious layers which entrap moisture and salts underneath [38].

4.3.4 Epoxies

The feasibility of using epoxies to consolidate stone is addressed by first briefly discussing their chemistry and then applications.

An epoxy consists of an epoxy resin and a curing agent which is actually a polymerization agent. Cure, i.e., polymerization, of an epoxy is initiated by mixing the epoxy resin with the curing agent. The epoxy resin is then converted into a hard thermosetting cross-linked polymer. The most commonly used epoxy resins are monomers of diphenylolpropane, called bisphenol A, and epichlorohydrin. Resins produced from these reactants are liquids, but are too viscous to penetrate stone deeply. Therefore, they are diluted with organic solvents. These epoxy resins are often cured using an amine curing agent. Their cure time can be adjusted by selecting a slowly or rapidly reacting curing agent and by controlling the curing temperature. The resulting cross-linked polymers have excellent adhesion to stone and concrete, and excellent chemical resistances. Two recommended sources for information on epoxies, such as their chemistry, curing, and applications are reference Nos. 132 and 133.

Gauri [134-135] developed a method to achieve deep penetration with viscous epoxy resins and at the same time avoid the formation of a sharp interface between the consolidated and untreated stone. First, specimens are soaked in acetone, then in a dilute solution of epoxy resin in acetone, followed by soaking in increasingly concentrated solutions. This method is feasible for small stone objects such as tombstones and statues, but would be too time consuming and expensive for stone structures.

Less viscous epoxy resins are available including diepoxybutane diglycidyl ether and butanediol diglycidyl ether [58]. Munnikendam [61] cured butanediol diglycidyl ether with alicyclic polyamines such as menthane diamine. However, the viscosity was still too high and he diluted the mixture with tetraethoxysilane and tetramethoxysilane. A complex reaction took place involving the epoxy resin, curing agent and solvent to produce a tough, glassy material. A white efflorescence also developed due to a reaction between the polyamine and carbon dioxide to form amine-carbonates [61, 137]. Formation of the aminecarbonates can be avoided by preventing carbon dioxide from coming in contact with the solution. Gauri [128, 136] observed that when low viscosity aliphatic epoxy resins were applied to calcareous stones, the reaction rates between the stones and carbon dioxide and sulfur dioxide were increased compared to the rates with untreated stones. He suggested that the increased reactivity could be caused by absorption of the gases by the epoxy polymer or by the polymer acting as a semipermeable film to the gases. In contrast, bisphenol A-based epoxy polymers were found to protect the stone from both carbon dioxide and sulfur dioxide.

The use of epoxies has been suggested for consolidating limestone [14, 128, 129], marble [134-139], and sandstone [61, 121] as well as for re-adhering large stone fragments to mass stone [1]. Moncrieff and Hempel [138] found that certain epoxies could encapsulate salts in marble, thereby preventing them from re-crystallizing. A large restoration project using epoxies for masonry consolidation is that of the Santa Maria Maggiore Church in Venice [140].

Similar to poly(methylmethacrylate), epoxies have produced brittle epoxy-impregnated concretes with high mechanical properties [65, 141,142]. The long-term effect on incorporating a brittle material in stone is not known, but could render a structure vulnerable to seismic shock, vibrations and thermal-dimensional effects.

Many types of epoxies have a tendency to chalk, i.e., to form a white powdery surface, when exposed to sunlight [132]. Therefore, epoxy should be removed from the surface of a treated stone before it cures.

4.3.5 Other Synthetic Organic Polymers

Other synthetic organic polymers studied as possible stone consolidants include polyester [38, 143], polyurethane [121], and nylon [77]. Polyester has been shown to decrease the porosity of stone substantially [143] and, therefore, may form an impervious layer which prevents the passage of entrapped moisture or salts [38]. Manaresi [121] and Steen [144] observed that polyurethanes were poor cementing agents. Steen [145] also found that polyurethane film gradually became brittle when exposed to sunlight. Similarly, DeWhite (77) found that nylon can produce a brittle film on the surface of stone.

4.4 Waxes

Waxes have been applied to stone for over 2,000 years. Vitruvius [146] described the impregnation of stone with wax in the first century B.C. A wax dissolved in turpentine was one of several materials applied to the decaying stone of Westminster Abbey between 1857 to 1859 [147].

Cleopatra's Needle (London, England) was treated with wax first in 1879 and several times since [148]. Kessler [149] found that paraffin waxes were effective in increasing the water repellency of stone. Waxes have also been found to be effective consolidants [25, 53, 54, 58]. For example, a paraffin wax increased the tensile strength of a porous stone from 1.06 MN/m^2 to 4.12 MN/m^2, while triethoxymethylsilane only increased it to 1.88 MN/m^2 [25, 53]. In addition, paraffin waxes are among the most durable stone conservation materials [16, 54] and can immobilize soluble salts [58].

Waxes have been applied to stone by applying the wax dissolved in organic solvents [16, 78, 148], by immersing a stone object in molten wax [58], or by applying molten wax to preheated stone [150]. If deep penetration is not achieved a nonporous surface layer may be formed causing the eventual spalling of the treated stone surface [78].

Major problems encountered in using waxes to conserve stone include their tendency to soften at high ambient temperatures [76], and to entrap dust and grime [2, 54, 58]. Wax applied to Cleopatra's Needle has gradually converted to a tarry substance which cannot be removed by ordinary washing methods. For example, a mixture of carbon tetrachloride, benzene and detergent was needed in 1947 to clean the Needle [148].

5. COMMENTS ON STONE CONSOLIDANTS

Although stone consolidants have been extensively used for over a century, their selection is still largely based on empirical considerations. If a consolidant appears to give acceptable results with one type of stone, it is often applied to other types of stone, without properly determining if the consolidant is compatible with them. Some of the factors affecting the performances of consolidants are known, such as depth of penetration and moisture transfer through consolidated stone. However, insufficient consideration has been given to equally important factors such as their consolidating abilities and the compatibility of their thermal-dimensional properties with stone. Finally, the long-term performances of consolidated stone of historic structures are rarely documented.

These considerations point to the inadequacy of the present state of stone consolidation and conservation technology. For example, stone consolidants should be selected on the basis of an understanding of the deterioration processes of stone, of the factors affecting the performances of consolidants, and of the compatibility of consolidants with specific stones. Presently, such information is often not available. Further, standard test methods and performance criteria should be developed to form the basis for selecting promising consolidants. Documentation of the performances of stone consolidants should be an integral part of each preservation or restoration program. Documentation of unsuccessful consolidation work is just as important as documenting successful work in that it assists other stone conservators in rejecting ineffective materials and methods.

This review clearly indicates that a perfect stone consolidant has not been developed and that many of the consolidants can do more harm to stone than natural weathering processes. Therefore, the general use of stone consolidants is open to question. In fact, the Commonwealth War Graves Commission, which is responsible for over one million headstones in Europe, has concluded [2] that no type of consolidant should be applied to headstones. This commission has over 50 years experience with the chemical treatment of stone. There are cases, however, in which the use of stone consolidants can be beneficial. The work by

Hempel and Moncrieff [115, 116, 131, 138, 139] has shown that decaying stone statues can be conserved by deep impregnation with certain stone consolidants. While statues and smaller objects can be removed to laboratories, surfaces thoroughly cleaned, freed from soluble salts, and all sides treated with a consolidant, such processes are not possible with massive stone structures. Risks involve in treating massive structures are, therefore, much greater. Consolidants might be used on structures of little historical or intrinsic value, and other cases where the benefits outweight the involved risks [2]. For example, consolidants could be applied to deteriorated stone to delay to a future time the need to replace it with new stone. (This approach could be dangerous if the consolidation measure is later regarded as "permanent"). Any permanent consolidation effort involving important historic stone structures should be carefully planned and carried out to minimize the risks. This includes making certain that moisture and soluble salts cannot become entrapped behind the treated stone. In addition, the compatibility of a consolidant with a specific stone should be determined with separated or isolated test specimens rather than using a historic structure as an experiment.

Stone conservators should be cautioned in the indiscriminate use of newly developed materials which have shown promise in accelerated laboratory tests. While these tests are useful in determining what materials are unacceptable, good results are not unequivocal proof that a material will have a lasting beneficial effect. Further, at present, we are not able to predict the durability of consolidated stone for even 20 to 30 years. Yet, unproven consolidants are being used on stone structure which have lasted for hundreds of years and which with rational conservation programs will last for many more years.

6. SUMMARY AND CONLCUSIONS

The main function of stone consolidating materials is to re-establish cohesion between particles of deteriorated stone. In addition to consolidation requirements, a good consolidant should meet performance requirements concerning durability, depth of penetration, effect on stone porosity, effect on moisture transfer, compatibility with stone, and effect on appearance. These have been termed "primary performance requirements" as they are considered to be generally invariable, i.e., they are essentially applicable to all stone consolidants regardless of the specific use. Secondary performance requirements are requirements which may be imposed in addition to the primary requirements because of specific problems encountered at certain structures. For example, to require a consolidant to immobilize soluble salts in a stone would be a secondary performance requirement.

Stone consolidants can be divided into four main groups, according to their chemistry. These groups are inorganic materials, alkoxysilanes, synthetic organic polymers, and waxes. Selection of what material to use depends on many factors including the type of stone to be consolidated, processes responsible for the deterioration of stone, degree of stone deterioration, the environment, amount of stone to be consolidated, and the importance of the stone structures. An universal consolidant does not exist because many of these factors will vary to some extent from structure to structure. Therefore, the preservation of each stone structure should be considered as an unique problem.

Few cases of long-term success with consolidating stone structures were disclosed in this review. Some apparent success has been achieved in consolidating small stone objects, such as statues, which can be treated in a laboratory. Consolidants should be used on massive stone structures only after an appraisal has been made which considers the risk involved, the benefits to be realized, and the probability of success.

7. ACKNOWLEDGMENTS

The author wishes to acknowledge the assistance of Mr. Henry Judd and Mr. Hugh Miller of the National Park Service, who provided information valuable to this review.

8. REFERENCES

1. G. Torraca, Treatment of Stone in Monuments. A Review of Principles and Processes, in the Conservation of Stone I, Proceeding of the International Symposium, Bologna, June 1975, pp. 297-315 (Centro per la Conservazione delle Sculture all'Aperto, Bologna, Italy, 1976).

2. W. H. Dukes, Conservation of Stone: Chemical Treatments, The Architects' Journal Information Library, pp. 433-438 (August 23, 1972).

3. W. H. Gutt, Conservation of Stone. The Next Steps, Stone Industries (London), May-June (1973).

4. G. Alessandrini, C. M. Del Fa, P. Rossi-Doria, M. Tabasso, and S. Vannucci, pp. 635-650, in reference 1.

5. J. Riederer, Further Progress in German Stone Conservation, pp. 369-385, in reference 1.

6. Preservation and Conservation: Principles and Practices (The Preservation Press, Washington, D.C., 1976)

7. G. A. Sleater, A Review of Natural Stone Preservation, NBSIR 74-444, National Bureau of Standards (Washington, D.C., 1973).

8. Stone Preservatives, Digest First Series No. 128, Building Research Station (Garston, England, 1963).

9. Colourless Treatments for Masonry, Digest 125, Building Research Station (Garston, England, 1971).

10. B. L. Clarke and J. Ashurst, Stone Preservation Experiments, Building Research Station (Garston, England, 1972).

11. Decay and Conservation of Stone Masonry, Digest 177, Building Research Station (Garston, England, 1975).

12. G. A. Sleater, Stone Preservatives: Method of Laboratory Testing and Preliminary Performance Criteria, NBS Technical Note 941, National Bureau of Standards (Washington, D.C., 1977)

13. S. Z. Lewin, The Preservation of Natural Stone, 1839-1965, An Annotated Bibliography, Art and Archaeology Technical Abstracts, Vol. 6 (No. 1), pp. 185-277 (1966).

14. E. M. Winkler, Stone: Properties, Durability in Man's Environment, 2nd edition (Springer-Verlag, New York, 1975).

15. R. J. Schaffer, The Weathering of Natural Building Stone, Building Research Special Report No. 18 (Garston, 1931); Reprinted 1972 by Building Research Station (Garston, England).

16. A. R. Warnes, Building Stones, Their Properties, Decay, and Preservation (Ernest Benn Ltd., London, 1926).

17. The Treatment of Stone, Proceedings of the Meeting of the Joint Committee for the Conservation of Stone, Bologna, October, 1971 (Centro per la Conservazione delle Sculture all'Aperto, Bologna, Italy, 1972).

18. The Conservation of Stone I, Proceedings of the International Symposium, Bologna, June 1975 (Centro per la Conservazione delle Sculture all'Aperto, Bologna, Italy, 1976).

19. Conservation of New Stone and Wooden Objects, New York Conference, June 1970 (The International Institute for Conservation of Historic and Artistic Works, London).

20. Deterioration and Protection of Stone Movements, International Symposium, June 1978 (Paris).

21. Decay and Preservation of Stone, Engineering Geology Case Histories No. 11, E. M. Winkler ed. (The Geological Society of America, Boulder, 1978).

22. D. Carroll, Rock Weathering (Plenum Press, New York, 1970).

23. F. C. Loughnan, Chemical Weathering of the Silicate Materials (Elsevier Publishing Co., 1969).

24. A. Arnold, Soluble Salts and Stone Weathering, pp. 133-135, in reference 18.

25. L. Arnold, D. B. Honeyborne and C. A. Price, Conservation of Natural Stone, Chemistry and Industry, pp. 345-347 (April 17, 1976).

26. J. Riederer, Stone Preservation in Germany, pp. 125-135, in reference 19.

27. W. H. Dukes, Conservation of Stone: Causes of Decay, The Architects' Journal Information Library, pp. 429-431 (August 23, 1972).

28. J. M. Garrido, The Portal of the Monastery of Santa Maria de Ripoll, Monumentum, Vol. 1, pp. 79-98 (1967).

29. W. H. Dukes, Conservation of Stone: Cleaning Natural Stone Buildings, The Architects' Journal Information Library, pp 497-504 (August 30, 1972).

30. E. M. Winkler, Stone Decay in Urban Atmospheres, pp. 53-58, in reference 21.

31. W. D. Keller, Progress and Problems in Rock Weathering Related to Stone Decay, pp. 37-46, in reference 21.

32. G. E. Likens, Acid Precipitation, Chemical and Engineering News, pp. 29-44 (November 22, 1976).

33. P. P. Hudec, Rock Weathering on the Molecular Level, pp. 47-51, in reference 21.

34. J. Lautridou and J. Ozouf, Relations Entre la Gelivite et les Proprietes Physiques (Porosite, Ascension Capillaire) des Roches Calcaires, Paper 3.3, in reference 20.

35. J. Hirschwald, Handbuch der Bautechnischen Gesteinsprüfung (Borntaeger; Berlin, 1912).

36. G. A. Bole, The Drain Tile Industry of Ohio, Clayworker, Vol. 90 (No. 4), pp. 286-288 (1928).

37. D. W. Kessler and W. H. Sligh, Physical Properties of the Principal Commercial Limestones Used for Building Construction in the United States, United States Bureau Technical Paper No. 34 (Government Printing Office, Washington, D.C., 1927).

38. H. A. La Fleur, The Conservation of Stone. A Report on the Practical Aspects of a UNESCO Course, October and November 1976, Venice, Italy (National Park Service, 1977).

39. L. Barcellona-Vero and M. Montesila, Mise en Evidence de l´Activite des Thiobacilles dans les Alterations des Pierres a Rome Identification de Certaines Souches, paper 4.1, in reference 20.

40. O. R. Lallement and S. Deruelle, Presence de Lichens sur les Monuments en Pierre: Nuisance au Protection, Paper 4.6., in reference 20.

41. S. B. Curri and A. Paleni, Some Aspects of the Growth of Chemolitotrophic Micro-Organisms on the Karnak Temples, pp. 267-278, in reference 18.

42. M. Godette, M. Post, and P. Campbell, Graffiti-Resistant Coatings: Methods of Test and Preliminary Selection Criteria, NBSIB 75-914, National Bureau of Standards (Washington, D.C., 1975).

43. M. Godette, M. Post, and P. Campbell, Graffiti Removers: Evaluation and Preliminary Selection Criteria, NBSIR 75-914, National Bureau of Standards (Washington, D.C., 1975).

44. J. R. Clifton, Protection of Reinforcing Bars with Organic Coatings, Materials Performance, Vol. 15 (No. 5), pp. 14-17 (1976).

45. Stainless Steel Reinforcement, BRE Information 99.78, Building Research Station (Garston, England, November 1978).

46. J. Riederer, No Decomposition of Stone by Air Pollution, Colloque International sur la Deterioration des Pierres en Oeuvre, 1st, La Rochelle (1972).

47. J. Riederer, Recent Advances in Stone Conservation in Germany, pp. 89-93, in reference 21.

48. Y. Efes, The Corrosion of Natural Stones at the Cathedral of Cologne, Centre Belge d'Etude et de Documentation des Eaux, No. 373, pp. 495-507 (December 1974).

49. Y. Efes and S. Luckat, Relations Between Corrosion of Sandstones and Uptakes Rates of Air Pollutants at the Cologne Cathedral, pp. 193-200, International Symposium on the Deterioration of Building materials (Athens, Greece, 1976).

50. I. E. Marsh, Stone Decay and Its Prevention (Basil Blackwell, Oxford, 1926).

51. G. Thomson and R. White, The pH of Rain and the Destruction of Alkaline Stone, Studies in Conservation, Vol. 19, pp. 190-191 (1974).

52. T. Stambolou, Conservation of Stone, pp. 119-124, in reference 19.

53. L. Arnold and C. A. Price, The Laboratory Assessment of Stone, pp. 695-704, in reference 18.

54. R. L. Gauri, J. A. Gwinn and R. K. Popli, Performance Criteria for Stone Treatment, pp. 143-152, in reference 49.

55. K. L. Gauri, D. J. Hagerty and C. R. Ullrich, Comparative Physical Properties of Weathered Impregnated and Unimpregnated Marble, Engineering Geology, Vol. 6, pp. 235-250 (1972).

56. G. Alessandrini, C. M. Del Fa, P. Rossi-Doria, M. Tabasso, and S. Vannucci, pp. 635-650, in reference 18.

57. R. A. Munnikendam, Preliminary Notes on the Consolidation of Porous Building Stones by Impregnation with Monomers, Studies in Conservation, Vol. 12 (No. 4), pp. 158-162 (1967).

58. C. A. Price, Stone Decay and Preservation, Chemistry in Britain, Vol. 11 (No. 9), pp. 350-353 (1975).

59. R. A. Munnikendam, Acrylic Monomer Sytems for Stone Impregnation, pp. 15-18, in reference 19.

60. G. Torraca, Brick, Adobe, Stone and Architectural Ceramics: Deterioration Processes and Conservation Practices, pp. 143-165, in reference 6.

61. R. A. Munnikendam, A New System for the Consolidation of Fragile Stone, Studies in Conservation, Vol. 18, pp. 95-97 (1973).

62. R. Wihr, Deep-Impregnation for Effective Stone-Protection, pp. 317-318, in reference 18.

63. K. L. Gauri, Preservation and Strengthening of Porous Solids, U.S. Patent 3,795,533 (1974).

64. L. E. Kukacha, A. Auskern, P. Colombo, J. Fontana, and M. Steinberg, Introduction to Concrete-Polymer Materials, Brookhaven National Laboratory; Available from National Technical Information Service, No. PB 241-691.

65. J. Clifton and G. Frohnsdorff, Polymer-Impregnated Concretes, pp. 174-196, in Cements Research Progress 1975 (American Ceramic Society, Columbus, Ohio, 1975).

66. E. M. Winkler and J. R. Clifton, Solvents for Adobe and Stone Preservatives, Paper 6.14, in reference 20.

67. E. Tammes and B. H. Vos, Vocht in Bouwconstructies, Bouwcentrum (Rotterdam, 1966).

68. J. R. Clifton, Preservation of Historic Adobe Structures-A Status Report, NBS Technical Note 934, National Bureau of Standards (Washington, D.C., 1977).

69. Standard Recommended Practice for Developing Short-Term Accelerated Tests for Prediction of the Service Life of Building Components and Materials, American Society for Test Materials (ASTM) Designation: E632-78.

70. J. R. Clifton, J. E. Fearn and E. D. Anderson, Polymer Impregnated Hardened Cement Pastes and Mortars, Building Science Series 83, National Bureau of Standards (Washington, D.C., 1976).

71. G. Biscontin and R. Pavan, A Consolidant for Stonework Conservation: Test and Experiences, paper 6.2, in reference 20.

72. G. Allesandrini, R. Peruzzi, P. Rossi-Doria and M. Tabasso, Control of the Behavior of the Two Epoxy Resins for "Stone" Treatments, Paper 6.1, in reference 20.

73. R. Munnikendam, private correspondence dated January 1, 1978 to J. R. Clifton.

74. Standard Method of Test for Absorption and Bulk Specific Gravity of Natural Building Stone, ASTM Designation: C97-47.

75. Standard Methods of Test for Water Vapor Transmission of Thick Materials, ASTM Designation: C355-64.

76. N. Heaton, The Preservation of Stone, Journal of the Royal Society of Arts, Vol. 70, pp. 129-139 (1921).

77. E. DeWhite, Soluble Nylon as Consolidation Agent for Stone, Studies in Conservation, Vol. 20 (No. 1), pp. 33-34 (1975).

78. A. P. Laurie and C. Ranken, The Preservation of Decaying Stone, Journal of the Society of the Chemical Industry, Vol. 37, pp. 137T-147T (1918).

79. G. Biscontin, S. Frascati, L. Marchesini, Colour Variations in Old Bricks and Stones as a Result of Consolidation with Resin, pp. 731-747, in reference 18.

80. Test for Instrumental Evaluation of Color Differences of Opaque Materials, ASTM Designation: D2244-68.

81. Test for Specular Gross, ASTM Designation: D523-67.

82. A. H. Munsell, Color Notation, Munsell Color Company, 18th revised edition (Baltimore, 1947).

83. E. W. Winkler, Simple Field Determination of Stone Colors, paper 3.9, in reference 20.

84. L. Arnold and C. A. Price, The Laboratory Assessement of Stone Preservatives, pp. 695-704, in reference 18.

85. A. P. Laurie, Building Materials, Oliver and Boyd Publishing Co. (Edinburgh, 1922).

86. J. Lehmann, Damage by Accumulation of Soluble Salts in Stonework, pp. 35-46, in reference 19.

87. D. S. Knopman, Conservation of Stone Artworks: Barely a Role for Science, Science, Vol. 190, pp. 1187-1188 (Dec. 19, 1975).

88. J. W. Mellor, A Comprehensive Treatise on Inorganic and Theoretical Chemistry, Vol. VI, Silicates (Longmans, Green and Co., London, 1925).

89. Encyclopedia of Chemical Reactions, Vol. VI (Reinhold Publishing Corp., New York, 1956.)

90. F. S. Barff, Stone Artificial Stone, Preserving Stone, Colouring, British Patent 2608 (1860); from reference 13.

91. L. Kessler, A Process for Hardening Soft Limestone by Means of the Fluosilicates of Insoluble Oxides, Comptes Rendus, Academie des Sciences, Institute de France, Vol. 96, pp. 1317-1319 (1883).

92. The Artificial Hardening of Soft Stone, Stone, Vol 34, pp. 365-366 (1913).

93. R. Wihr and G. Steenken, On the Preservation of Monuments and Works of Art with Silicates, pp. 71-75, in reference 19.

94. B. G. Shore, Stones of Britain (Leonard Itill Books Ltd., London, 1957).

95. B. Penkala, The Influence of Surface Protecting Agents on the Technical Properties of Stone, Ochrona Zabytrow, Vol 17 (No. 1), pp. 37-43 (1964).

96. A. R. Powys, Repair of Ancient Buildings (J. M. Dent & Sons, Ltd., London, 1929).

97. W. Linke, Solubilities of Inorganic and Metals Organic Compounds, Vol. I (Van Nostrand, Princeton, 1958).

98. H. G. Lloyd, Hardening Stone and Earth, British Patent 441,568 (1934).

99. V. Mankowsky, The Weathering of Our Large Monuments, Die Denkmalpflege Vol. 12, (No. 1), pp. 51-54 (1910); from reference 13.

100. Breathing New Life into the Statues of Wells, New Scientist, pp. 754-756 (22/29 December 1977).

101. A. H. Church, Stone, Preserving and Colouring; Cements, British Patent 220 (1862); from reference 13.

102. A. H. Church, Treatment of Decayed Stone-work in the Chapter House, Westminster Abbey, Journal of the Society of Chemical Industry, Vol. 23, pg. 824 (1904).

103. A. H. Church, Conservation of Historic Buildings and Frescoes, Proceedings of the Meetings of the Members, Royal Institution of Great Britain, Vol. 18, pp. 597-608 (1907).

104. S. Z. Lewin, The Conservation of Limestone Objects and Structures in Study of Weathering of Stone, Vol. 1 (International Council of Monuments and Sites, Paris, 1968).

105. E. V. Sayre, Direct Deposition of Barium Sulfate from Homogeneous Solution Within Porous Stone, pp. 115-118, in reference 19.

106. G. G. Scott, Process as Applied to Rapidly-Decayed Stone in Westminster Abbey, The Builder (London), Vol. 19, pp. 105 (1861); from reference 13.

107. G. Zava, B. Badan and L. Marchesini, Structural Regeneration by Induced Mineralization of the Stone of Eraclea (Agrigento) Theatre, pp. 387-399, in reference 18.

108. S. Z. Lewin and N. S. Baer, Rationale of the Barium Hydroxide-Urea Treatment of Decayed Stone, Studies in Conservation, Vol. 19, pp. 24-35 (1974).

109. C. A. Price, Research on Natural Stone at the Building Research Establishment, Natural Stone Directory (1977).

110. Bosch, Use of Silicones in Conservation of Monuments, pp. 21-26, First International Symposium on the Deterioration of Building Stones (1972).

111. H. Weber, Stone Renovation and Consolidation Using Silicones and Silicic Esters, pp. 375-385, in reference 18.

112. Preserving Building Stone, BRE News, Vol. 42, pp. 10-11 (Winter, 1977).

113. J. Taralon, C. Jaton, and G. Orial, Etat des Recherches Effectuees en France sur les Hydrofuges, Studies in Conservation, Vol. 20, pp. 455-476 (1975).

114. J. Riederer, Die Echaltung Ägyptischer Baundenkmäler, Maltechnik Restauro, Vol. 74 (No. 1), pp. 43-52 (1974).

115. A Moncrieff, the Treatment of Deteriorating Stone with Silicone Resins: Interim Report, Studies in Conservation, Vol. 21, pp. 179-191 (1976).

116. K. Hempel and A. Moncrieff, Report on Work since Last Meeting in Bologna, October 1971, pp. 319-339, in reference 18.

117. A. P. Laurie, Preservation of Stone, U.S. Patent 1,607,762 (1926).

118. H. D. Cogan and C. A. Setterstrom, Ethyl Silicates, Industrial Engineering Chemistry, Vol. 39, pp. 1364-8 (1947).

119. H. D. Cogan and C. A. Setterstrom, Chemical Engineering News, Vol. 24, pp. 2499-2501 (1946).

120. H. Marschner, Application of Salt Crystallization Test to Impregnated Stones, paper 3.4, in reference 20.

121. R. Snethlage and D. D. Klemm, Scanning Electron Microscope Investigations on Impregnated Sandstones, paper 5.7, in reference 20.

122. R. Rossi-Manaresi, Treatments for Sandstone Consolidation, pp. 547-571, in reference 18.

123. M. B. Caroe, Wells Cathedral, The West Front Conservation Programme, Interim Report on Aims and Techniques (June, 1977).

124. M. Steinberg et. al., Concrete Polymer Materials, First Tropical Report, Brookhaven National Laboratory Report No. BNL-50134 (1968).

125. L. E. Kukacka, et. al., Concrete-Polymer Materials, Fifth Topical Report, Brookhaven National Laboratory Report No. BNL-50390 (1973).

126. E. Dahl-Jorgensen and W. F. Chen, Stress-Strain Properties of Polymer Modified Concrete, Fritz Engineering Laboratory Report No. FEL 390.1 (Lehigh University, Pennsylvania, 1973).

127. R. B. Seymour, Introduction to Polymer Chemistry (McGraw-Hill, Inc., 1971).

128. K. L. Gauri, P. Tanjaruphan, M. A. Rao and T. Lipscomb, Reactivity of Treated and Untreated Marble in Carbon Dioxide Atmospheres, Transactions of the Kentucky Academy of Science, Vol. 38 (No. 1-2), pp. 38-44 (1977).

129. K. L. Gauri and M. V. A. Rao, Certain Epoxies, Fluorocarbon-Acrylics and Silicones as Stone Preservatives, pp. 73-80, in reference 21.

130. Usefulness of Some Vinyl Polymers in the Conservations of Monuments, Ochrona Zabytkow, Vol 14 (No. 3-4), pp. 81-92 (1961); from reference 13.

131. K. F. B. Hempel, Notes on the Conservation of Sculptue, Stone, Marble, and Terracotta, Studies in Conservation, Vol. 13, pp. 34-44 (1968).

132. H. Lee and K. Neville, Handbook of Epoxy Resins (McGraw-Hill Book Company, 1967).

133. W. G. Potter, Uses of Epoxy Resins (Chemical Publishing Company, New York, 1975).

134. K. L. Gauri, Cleaning and Impregnation of Marble, in reference 17.

135. K. L. Gauri, Improved Impregnation Technique for the Preservation of Stone Statuary, Nature, Vol. 228 (No. 5274), pg. 882 (Nov. 28, 1970).

136. K. L. Gauri, Efficiency of Epoxy Resins as Stone Preservatives, Studies in Conservation, Vol. 19, pp. 100-101 (1974).

137. G. Marinelli, Use of an Epoxy Aliphatic Resin in the Consolidation of Porous Building Materials Having Poor Mechanical Properties, pp. 573-591, in reference 18.

138. A. Moncrieff, Work on the Degeneration of Sculptural Stone, pp. 103-114, in reference 19.

139. K. Hempel and A. Moncrieff, Summary of Work on Marble Conservation at the Victoria and Albert Museum Conservation Department up to August 1971, in reference 17.

140. Epoxy Resins Saves Venice Church, Corrosion Prevention and Control, pp. 12-13 (October, 1974).

141. D. A. Whiting, P. R. Blankenhorn and D. E. Kline, Effect of Hydration on the Mechanical Properties of Epoxy Impregnated Concrete, Cement and Concrete Research, Vol. 4 (No. 3), pp. 467-476 (1964).

142. D. A. Whiting, P. R. Blankenhorn and D. E. Kline, Mechanical Properties of Epoxy Modified Concrete, Journal of Testing and Evaluation, Vol. 2 (No. 1), pp. 44-49 (1974).

143. M. Kranz, l'Evaluation de l'Etat de Conservation de la Pierre et de l'Efficacite des Traitements, pp. 443-453, in reference 18.

144. C. R. Steen, Some Recent Experiments in Stabilizing Adobe and Stone, pp. 59-64, in reference 19.

145. C. Steen, An Archaelogist's Summary of Adobe, El Palacio, Vol. 77 (No. 4), pp. 29-38 (1971)

146. Vitruvius, de Architecturia, I.X.; from reference 58.

147. G. G. Scott, Process as Applied to Rapidly-Decayed Stones in Westminster Abbey, The Builder, Vol. 19 (No. 941), pp. 105 (1861); from reference 13.

148. S. G. Burgess and R. J. Schaffer, Cleopatra's Needle, Chemistry and Industry, pp. 1026-1029 (1952).

149. D. W. Kessler, Exposure Tests on Colourless Materials, Technological Papers of the Bureau of Standards, No. 248.

150. The Preservation of Ruins, Literary Digest, pp. 94-95 (July 16th, 1910).

NBS-114A (REV. 9-78)

U.S. DEPT. OF COMM.
BIBLIOGRAPHIC DATA SHEET

1. PUBLICATION OR REPORT NO. NBS TN 1118

2. Gov't. Accession No.

3. Recipient's Accession No.

4. TITLE AND SUBTITLE

Stone Consolidating Materials--
A Status Report

5. Publication Date May 1980

6. Performing Organization Code

7. AUTHOR(S)

James R. Clifton

8. Performing Organ. Report No.

9. PERFORMING ORGANIZATION NAME AND ADDRESS

NATIONAL BUREAU OF STANDARDS
DEPARTMENT OF COMMERCE
WASHINGTON, DC 20234

10. Project/Task/Work Unit No.

11. Contract/Grant No.

12. SPONSORING ORGANIZATION NAME AND COMPLETE ADDRESS *(Street, City, State, ZIP)*

National Park Service
U. S. Department of Interior
Washington, D.C. 20240

13. Type of Report & Period Covered Final

14. Sponsoring Agency Code

15. SUPPLEMENTARY NOTES

☐ Document describes a computer program; SF-185, FIPS Software Summary, is attached.

16. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)*

Information on types of stone consolidating materials, their performances, and uses are critically reviewed. Processes responsible for the deterioration of stone and criteria for selecting stone consolidants are also reviewed. The main function of stone consolidants is to reestablish the cohesion between particles of deteriorated stone. In addition, a good consolidant should meet performance requirements concerning durability, depth of penetration, effect on stone porosity, effect on moisture transfer, compatibility with stone, and effect on appearance.

Stone consolidants can be divided into four main groups, according to their chemistry. These groups are inorganic materials, alkoxysilanes, synthetic organic polymers, and waxes. Epoxies, acrylics, and alkoxysilanes are currently the most commonly used consolidating materials. Certain waxes have been found to have excellent consolidating abilities. Waxes, however, tend to soften and to accumulate grime and dust. Inorganic materials and some organic polymers have a tendency to form shallow, brittle layers near the surface of stone. These treated layers often separate from the untreated stone. Alkoxysilanes have excellent penetrabilities and are considered by some stone conservators as the most promising consolidating materials. However, a universal consolidant does not exist and many factors must be considered in selecting a consolidant for a specific stone structure.

17. KEY WORDS *(six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)*

Conservation; consolidating materials; deterioration of stone; preservation; stone; stone consolidation.

18. AVAILABILITY ☒ Unlimited

☐ For Official Distribution. Do Not Release to NTIS

☒ Order From Sup. of Doc., U.S. Government Printing Office, Washington, DC 20402, ____________

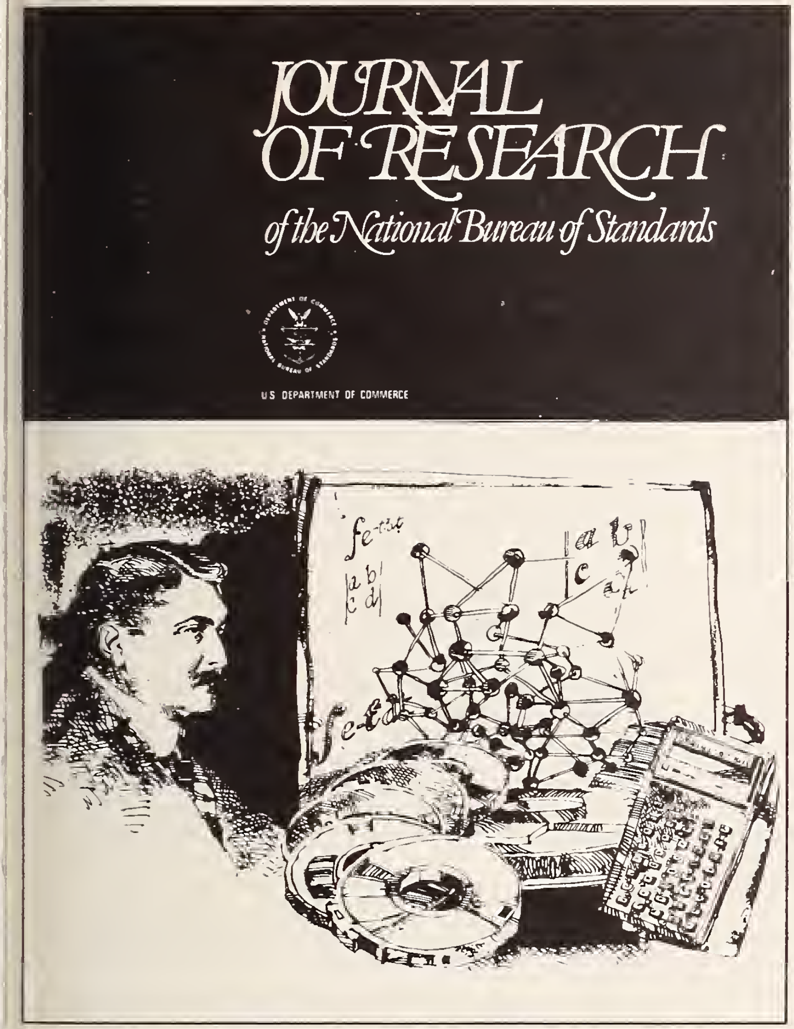
☐ Order From National Technical Information Service (NTIS), Springfield, VA. 22161

19. SECURITY CLASS (THIS REPORT) UNCLASSIFIED

20. SECURITY CLASS (THIS PAGE) UNCLASSIFIED

21. NO. OF PRINTED PAGES 52

22. Price $3.50



Subscribe now— The new National Bureau of Standards Journal

The expanded Journal of Research of the National Bureau of Standards reports NBS research and development in those disciplines of the physical and engineering sciences in which the Bureau is active. These include physics, chemistry, engineering, mathematics, and computer sciences. Papers cover a broad range of subjects, with major emphasis on measurement methodology, and the basic technology underlying standardization. Also included from time to time are survey articles on topics closely related to the Bureau's technical and scientific programs. As a special service to subscribers each issue contains complete citations to all recent NBS publications in NBS and non-NBS media. Issued six times a year. Annual subscriptions: domestic $17.00; foreign $21.25. Single copy, $3.00 domestic; $3.75 foreign.

- Note: The Journal was formerly published in two sections: Section A "Physics and Chemistry" and Section B "Mathematical Sciences."

Subscription Order Form
Enter my subscription to NBS Journal of Research at $17.00. Add $4.25 for foreign mailing. No additional postage is required for mailing within the United States or its possessions. (SJRA—File Code 2N)

Send Subscription to:

Name-First, Last

Company Name or Additional Address Line

Street Address

City | State | Zip Code

(or) COUNTRY

PLEASE PRINT

Credit Card Orders Only
Total charges $ .... Fill in the boxes below.
Credit Card No.
VISA master charge
Master Charge Interbank No.
Expiration Date Month/Year

☐ Remittance Enclosed. Domestic: Check or money order. Foreign: International money order, draft on an American or Canadian Bank, or by UNESCO coupons, Made payable to the Superintendent of Documents.

☐ Charge to my Deposit Account No.

MAIL ORDER FORM TO:
Superintendent of Documents
Government Printing Office
Washington, D.C. 20402

☆ U.S. GOVERNMENT PRINTING OFFICE : 1980 O-311-055 (83)

NBS TECHNICAL PUBLICATIONS

PERIODICALS

JOURNAL OF RESEARCH—The Journal of Research of the National Bureau of Standards reports NBS research and development in those disciplines of the physical and engineering sciences in which the Bureau is active. These include physics, chemistry, engineering, mathematics, and computer sciences. Papers cover a broad range of subjects, with major emphasis on measurement methodology and the basic technology underlying standardization. Also included from time to time are survey articles on topics closely related to the Bureau's technical and scientific programs. As a special service to subscribers each issue contains complete citations to all recent Bureau publications in both NBS and non-NBS media. Issued six times a year. Annual subscription: domestic $17; foreign $21.25. Single copy, $3 domestic; $3.75 foreign.

NOTE: The Journal was formerly published in two sections: Section A "Physics and Chemistry" and Section B "Mathematical Sciences."

DIMENSIONS/NBS—This monthly magazine is published to inform scientists, engineers, business and industry leaders, teachers, students, and consumers of the latest advances in science and technology, with primary emphasis on work at NBS. The magazine highlights and reviews such issues as energy research, fire protection, building technology, metric conversion, pollution abatement, health and safety, and consumer product performance. In addition, it reports the results of Bureau programs in measurement standards and techniques, properties of matter and materials, engineering standards and services, instrumentation, and automatic data processing. Annual subscription: domestic $11; foreign $13.75.

NONPERIODICALS

Monographs—Major contributions to the technical literature on various subjects related to the Bureau's scientific and technical activities.

Handbooks—Recommended codes of engineering and industrial practice (including safety codes) developed in cooperation with interested industries, professional organizations, and regulatory bodies.

Special Publications—Include proceedings of conferences sponsored by NBS, NBS annual reports, and other special publications appropriate to this grouping such as wall charts, pocket cards, and bibliographies.

Applied Mathematics Series—Mathematical tables, manuals, and studies of special interest to physicists, engineers, chemists, biologists, mathematicians, computer programmers, and others engaged in scientific and technical work.

National Standard Reference Data Series—Provides quantitative data on the physical and chemical properties of materials, compiled from the world's literature and critically evaluated. Developed under a worldwide program coordinated by NBS under the authority of the National Standard Data Act (Public Law 90-396).

NOTE: The principal publication outlet for the foregoing data is the Journal of Physical and Chemical Reference Data (JPCRD) published quarterly for NBS by the American Chemical Society (ACS) and the American Institute of Physics (AIP). Subscriptions, reprints, and supplements available from ACS, 1155 Sixteenth St., NW, Washington, DC 20056.

Building Science Series—Disseminates technical information developed at the Bureau on building materials, components, systems, and whole structures. The series presents research results, test methods, and performance criteria related to the structural and environmental functions and the durability and safety characteristics of building elements and systems.

Technical Notes—Studies or reports which are complete in themselves but restrictive in their treatment of a subject. Analogous to monographs but not so comprehensive in scope or definitive in treatment of the subject area. Often serve as a vehicle for final reports of work performed at NBS under the sponsorship of other government agencies.

Voluntary Product Standards—Developed under procedures published by the Department of Commerce in Part 10, Title 15, of the Code of Federal Regulations. The standards establish nationally recognized requirements for products, and provide all concerned interests with a basis for common understanding of the characteristics of the products. NBS administers this program as a supplement to the activities of the private sector standardizing organizations.

Consumer Information Series—Practical information, based on NBS research and experience, covering areas of interest to the consumer. Easily understandable language and illustrations provide useful background knowledge for shopping in today's technological marketplace.

Order the **above** *NBS publications from: Superintendent of Documents, Government Printing Office, Washington, DC 20402.*

Order the **following** *NBS publications—FIPS and NBSIR's—from the National Technical Information Services, Springfield, VA 22161.*

Federal Information Processing Standards Publications (FIPS PUB)—Publications in this series collectively constitute the Federal Information Processing Standards Register. The Register serves as the official source of information in the Federal Government regarding standards issued by NBS pursuant to the Federal Property and Administrative Services Act of 1949 as amended, Public Law 89-306 (79 Stat. 1127), and as implemented by Executive Order 11717 (38 FR 12315, dated May 11, 1973) and Part 6 of Title 15 CFR (Code of Federal Regulations).

NBS Interagency Reports (NBSIR)—A special series of interim or final reports on work performed by NBS for outside sponsors (both government and non-government). In general, initial distribution is handled by the sponsor; public distribution is by the National Technical Information Services, Springfield, VA 22161, in paper copy or microfiche form.

BIBLIOGRAPHIC SUBSCRIPTION SERVICES

The following current-awareness and literature-survey bibliographies are issued periodically by the Bureau:

Cryogenic Data Center Current Awareness Service. A literature survey issued biweekly. Annual subscription: domestic $25; foreign $30.

Liquefied Natural Gas. A literature survey issued quarterly. Annual subscription: $20.

Superconducting Devices and Materials. A literature survey issued quarterly. Annual subscription: $30. Please send subscription orders and remittances for the preceding bibliographic services to the National Bureau of Standards, Cryogenic Data Center (736) Boulder, CO 80303.

U.S. DEPARTMENT OF COMMERCE
National Bureau of Standards
Washington, D.C. 20234