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The Effects of Herbicides on Masonry

James E. Fearn

Structures and Materials Division
Center for Building Technology
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National Bureau of Standards
Washington, D.C. 20234

Final Report

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THE EFFECTS OF HERBICIDES ON MASONRY

James E. Fearn

Abstract

In preserving historic structures, the control of obnoxious vegetation is a serious problem. To deal with this problem, a number of organic herbicides have been developed by industry. The efficacy of herbicides in the control of plant life has been studied to a great degree, but heretofore, very little has been reported about the possible effects of these chemical plant killers on the materials they are supposed to protect. In this work, an exhaustive survey of pertinent literature was undertaken. Obtaining very little specific information from literature, a correlation has been drawn between the effects on masonry of materials similar in chemistry to herbicides and the effects that would be expected from the herbicides themselves. Methods for checking the validity of conclusions are suggested.

Key words: acidic, alkaline, degradation, herbicide, historic structures, masonry.

1. INTRODUCTION

1.1 BACKGROUND

The destructive effects of vegetation on artistic or historic structures are a serious concern to those entrusted with their care. This is true both in this country [1][1] and abroad [2-5]. Damage may be caused by such simple plants as algae, fungi, lichens, and mosses, or by the more sophisticated flora commonly referred to as weeds. The damage may result from water retention by foliage, the weakening of mortar joints through penetration and proliferation of roots [2], or through chemical attack by certain secretions of the plant.

The usual method of combatting pernicious vegetation is by repeated cutting and clearing but in areas of alternate bright sunlight and heavy rainfall, where the growth rate of plants is particularly high, chemical control may be competitive with the older methods. Unfortunately, uncertainty about the effects of the chemicals on masonry makes difficult the assessment of the risk of using herbicides while it is known that careful cutting and clearing is safe and effective. The National Park Service is interested in the use of herbicides to help in the difficult and costly task of controlling vegetation in the immediate vicinity of a series of forts built along the Gulf of Mexico. Because of the uncertainty of the long-term effects of herbicides on the masonry of these forts the National Park Service asked the National Bureau of Standards to carry out the survey covered in this report.

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[1] Numbers in brackets refer to the literature references listed at the end of this paper.

1.2 OBJECTIVES AND SCOPE

1.2.1 Objectives

1. To survey and assess the state of knowledge regarding the effects of herbicides on masonry structures and to present preliminary recommendations about the use of herbicides.
2. If deemed necessary in the light of the results to prepare a comprehensive research plan for evaluating the effects of herbicides on masonry structures.

1.2.2 Scope

While the study was begun to provide information concerning the effects of commercially available herbicides on the masonry of the Gulf Coast forts, general considerations of the effects of herbicides on masonry of all types have been included in the hope that they will assist all who are faced with the task of protecting artistic or historic masonry structures from the destructive effects of vegetation.

This report is organized in such fashion that the range of masonry materials is discussed first (Chapter 2), followed by a brief review of what is known about the degradation of these materials or their constituents by chemicals (Chapter 3). Next, the compositions and natures of the most common types of herbicides are reviewed (Chapter 4), and the possible effects of herbicides on masonry materials considered from the chemistry of these materials (Chapter 5). In Chapter 6, preliminary criteria for selecting herbicides for use on masonry are presented along with commentaries on research which may be carried out in improving the technical bases for the criteria and reducing them to their most conveniently useful form. Finally, a summary of the report with recommendations concerning the use of herbicides on artistic and historic masonry structures is given in Chapter 7.

2. MASONRY MATERIALS

2.1 MASONRY UNITS

Masonry units are of many kinds, differing widely in appearance and durability. They include many types of building stones (table 1) [14], clay brick, clay tile, terra cotta, cinder block, concrete block, adobe, etc. Clay brick, which was developed by potters prior to the building of King Soloman's temple [6], during the tenth century before Christ, may vary widely in durability and other aspects of performance because of differences in raw materials and manufacturing processes. This is true of manufactured masonry units in general. Variation in building stones results from differences in the natural processes by which they were formed. At the time of the building of the Gulf Coast forts in the mid-nineteenth century, the lack of consistency in the quality of brick was a major problem [7]. It is therefore not judicious to consider modern clay brick an adequate model for the brick used in constructing the forts. The ability of brick and other clay products to resist weathering depends to a considerable extent on their porosities, which vary widely depending on the specific nature of the original clay and the method of fabrication [6] (see table 1).

An excellent book by McKee [17] traces beautifully the development of the brick industry in America from 1610 to 1758. Norton [18] also deals with such important aspects of brick fabrication as the construction of kilns and critical temperatures needed in producing bricks of desired quality.

Table 1

Mineral Composition of Masonry Materials

Masonry Material	Porosity (%)	Typical Mineral Composition	Ref.
Granite	0.5-1.5	Feldspars, quartz, hornblende, mica	[14]
Gabbro	0.1-0.2	Hornblende, feldspar, plagioclase, mica	
Basalt	0.1-1.0	Hornblende, feldspars	
Porphyry	0.5-1.5	Feldspar, hornblende	
Limestone	5.0-20.0	Calcite, clay minerals	
Sandstone	5.0-25.0	Sand, quartz, feldspar, calcite	
Dolomite	1.0-5.0	Calcium carbonate, magnesium carbonate	
Travertine	5.0-20.0	Calcite, various other metallic compounds	
Shale	10.0-30.0	Mica, quartz, calcite, clay, metalic oxides	
Slate	0.1-0.5	Mica, quartz, calcite	
Marble	0.5-2.0	Calcite, mica, quartz	
Quartzite	0.1-0.5	Quartz, mica, sand	
Gneiss	0.5-1.5	Feldspar, quartz, hornblende	
Clay Brick	23.0-40.0	Clay or shale (largely aluminum silicate)	[9]
Clay Tile	23.0-40.0	Clay or shale, silica, metallic oxides	
Terra Cotta	23.0-40.0	Clay, silica, metallic oxides	
Hardened Portland Cement	5.0-40.0	Hydrates of calcium silicate, calcium aluminates, calcium aluminoferrite, calcium oxide, calcium sulphates, water	

Cinder Block	5.0-20.0	Hardened hydrated portland cement, quartz, cinders	
Concrete Block	1.0-10.0	Hardened portland cement, quartz gravel	
Lime Mortar	1.0-10.0	Calcium oxide, calcium hydroxide, calcium carbonate, quartz	[19]
Portland Cement Mortar	1.0-10.0	Calcium silicate hydrate, calcium hydroxide, sand, water (occasionally lime)	

Note: Porosities for cement products indicate only orders of magnitude.

2.2 MORTARS

Among others, Taylor [19], Brunauer and Copeland [8, 19] have dealt exhaustively with the subject of portland cement and the various hydrates produced when it combines with water. Portland cement and its various modifications grew out of the discovery patented by John Aspdin [8], an English bricklayer, in 1824. This material has largely replaced most of the earlier cementitious materials used in mortars.

Earlier mortars based on lime (calcium oxide) have been used for more than 5000 years, since they were used by the Egyptians in the construction of the pyramids. Much later the Romans combined lime paste with volcanic ash to produce mortar which they called hydraulic lime, since this mortar when mixed with water give insoluble cementitious products. Mortars based on lime were still in common use during the planning and early phases of the construction of the Gulf Coast forts [7]. Combinations of slaked lime and portland cement are still employed for some masonry purposes.

The chemical natures of constituents of hardened mortars and other masonry materials which include the elements of hardened mortars are indicated in table 1. Porosities given for these materials indicate only orders of magnitude since in Portland cement mortars porosity varies widely with the water-cement ratio and the age of the mix. Possible mechanisms of interactions between these mortars and herbicides will be discussed in the following chapter.

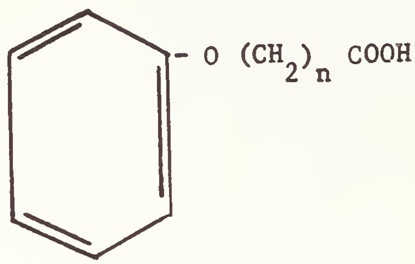
3. DEGRADATION OF MASONRY MATERIALS

The rate of attack of masonry materials of chemical agents depends on a number of factors, among them the chemistry of the masonry material, the chemistry of the attacking substance and the amount of surface exposed to attack. With mortar this surface is determined to a large extent by its porosity. Other factors being equal, the rate of attack varies directly with porosity. For the masonry materials listed in table 1, the decreasing order of resistance to chemical attack is roughly as follows: quartz, feldspar, calcite, calcium silicate hydrate, calcium hydroxide. The principal ingredients of mortar are sand, hydrated calcium silicates and calcium hydroxide; because of its composition and its high porosity, mortar is more vulnerable to chemical attack than most masonry units. Portland cement mortar and its predecessor, lime mortar, are so stable at ambient temperatures and in dry environments that samples of lime mortar known to be 4500 years old are still in good condition [8]. Their solubility in water is low but not negligible, and attack by strong alkali is a potential danger. Ammonium compounds tend to be damaging to hardened portland cement and lime products, as are materials which produce the sulfate ion. All acids have a deleterious effect on concrete and mortar, the effect varying roughly with the strength of the acid [9, 10, 11]. Fats and oils which may be saponified in the presence of calcium hydroxide or which may undergo hydrolysis or oxidation to produce acids will attack portland cement and lime mortars. The presence of carbon dioxide or sulfur dioxide in substantially higher quantities than are normal in air accelerates the decomposition of portland cement and lime mortars [9, 10]. Kleinlogel [10] presents an extensive list of substances and the effects of these substances on masonry materials. The Structural Clay Products Institute, which has been succeeded by the Brick Institute of America, published a Technical Note on the effects of various materials on mortars which is less extensive than the book by Kleinlogel but is more recent in its date of publication [9]. Masonry units which contain large amounts of calcite, e.g., limestones, marble and a number of sandstones, are subject to acid attack. Those containing largely quartz or other forms of silica, e.g., granite, porphyry, and gneiss are considerably less so. Fired clay brick is particularly resistant to attack by chemicals. Unfired clay adobe and bricks are not usually subject to chemical attack but over long periods of time are eroded by running water.

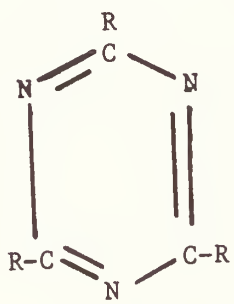
4. HERBICIDES

Kearney, Kaufman, et al. [12] have separated herbicides into eleven categories for purposes of studying their degradative reactions. A list of these categories with the basic structure for each follows.

a. Phenoxy alkanoic Acids

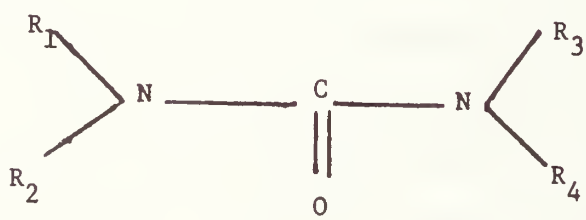


b. S-Triazines

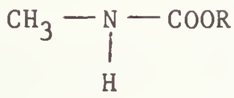


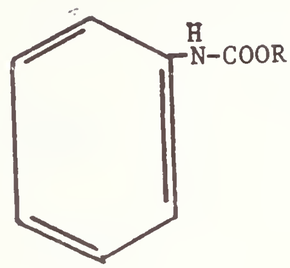
R = CH_3, C_2H_5, etc.

c. Substituted Ureas



d. Methyl Carbamates and Phenyl Carbamates

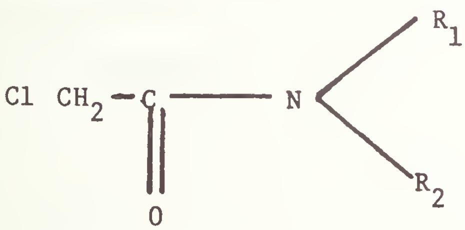




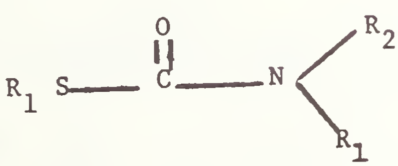
e. Chlorinated Aliphatic Acids

$CX_3\ (CXY)_n\ COOR$ where X = H,Cl; Y = Cl,H; and n = 0, 1, 2, ...

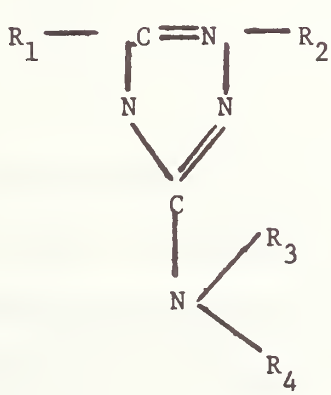
f. Chloroacetamides



g. Thiolcarbamates



h. Amitrole[2]



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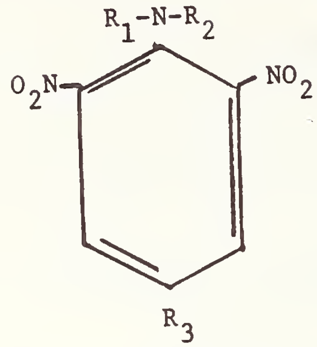
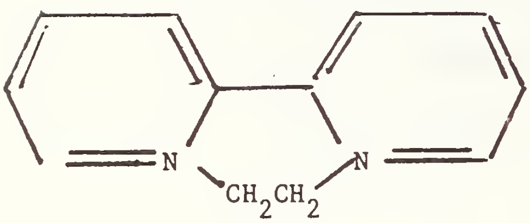
[2] The use of commercial designations is a convenience in identifying particular chemicals; it does not imply endorsement of the material by the National Bureau of Standards.

i. Trifluralin and Related Compounds

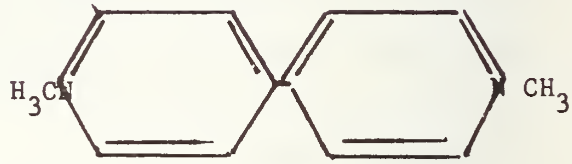


where $R_3 = CF_3$, CH_3, SO_2CH_3, etc.

j. Diquat and Paraquat

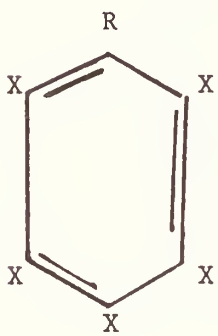


Diquat



Paraquat

k. Benzoic Acid Herbicides



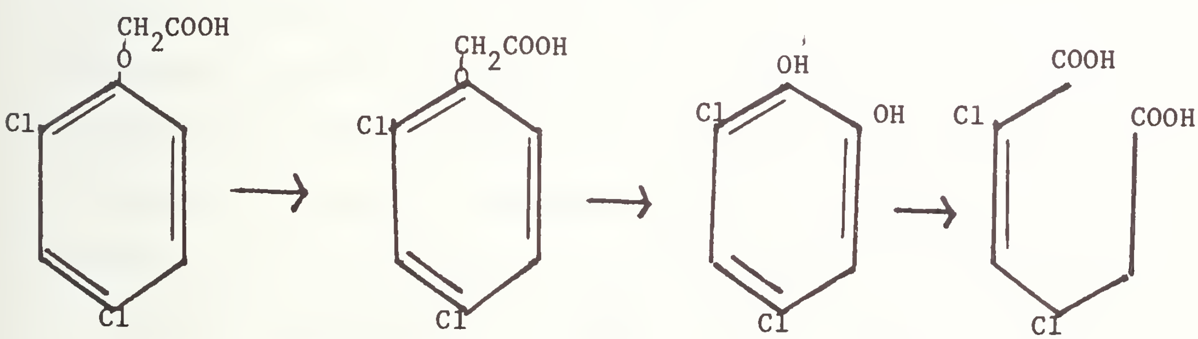
where R = COOH, CN, CONH, $CSNH_2$, etc.

and X = Cl, OH, OCH_3, NH_2, etc.

The Farm Chemicals Handbook [20] gives pertinent data on most commercial members of all eleven of these categories. Given the commercial name, this volume will present the chemical name and formula and other commercial names, its chemical action and properties, toxicity, applications, formulations, combinations, limitations, etc.

5. DISCUSSION OF HERBICIDAL EFFECTS ON MASONRY

It must be understood that the formulas illustrated represent families of compounds rather than specific materials. Also important is the fact that many commercial herbicides are mixtures of members of two or more of these categories. The commercial preparation Tordon (R) 101R is, for example, a mixture of the triisopropanol amine salt of 2, 4 - dichlorophenoxy acetic acid (category a) and the triisopropanol amine salt of 3, 5, 6 - trichloropicolinic acid. Since the amine salts of these two carboxylic acids are weakly alkaline in aqueous solution (pH more than 7), it is unlikely that damage would occur to masonry if this material were used to control vegetation but care must be taken to consider the degradation products of these materials. Consider, for example, the oxidative decomposition of 2, 4 - dichlorophenoxy acetic acid under the influence of certain microbes or through certain other methods of oxidaton.



I II III IV

$\rightarrow CO_2 + H_2O + Cl^-$.

As indicated, the phenoxyacetic acid (I) is converted to the hydroxy-phenoxy acetic acid (II) which is further oxidized dichloro catechol (III) the β, β' dichloro-muconic acid (IV) and, finally, carbon dioxide, chloride ion and water.

Although with Tordon (R) 101R the amine may completely neutralize the acetic acid group of the original molecule, considerable acidity may develop as the ring portion of the molecules changes gradually to form muconic acid. When total decomposition occurs, carbon dioxide and chloride ion are produced both of which are damaging to cementitious materials when the quantity is sufficiently large and water is present [10, 16]. The present level of understanding does not include the rates of the reactions outlined above. It is therefore, impossible to determine the maximum concentration of corrosive elements at any given time after masonry has been exposed to this herbicide. A generalization may be from Kearney's report [12] and from classical organic chemistry that oxidation of herbicides, as of organic substances in general, generally produces materials of greater acidity than the original material. Hydrolyses catalyzed by enzymes also increase acidity. Fats or oils are saponified, through a metathetical reaction in which the acid component of the fat or oil attacks the calcium of the cementitious material. One may thus conclude, in the absence of specific information to the contrary, that any acid that is water soluble, even though only slightly so, or any material which may oxidize or hydrolyze to produce acid, must be regarded as a possible threat to the masonry. One must consider also the possible corrosive effect of either acid or alkaline solutions of herbicides on any metallic substance intimately associated with the masonry.

6. LITERATURE SURVEY AND PRELIMINARY CRITERIA FOR EVALUATING HERBICIDES

6.1 LITERATURE SURVEY OF EFFECTS OF HERBICIDES ON MASONRY

An intense computer search through all pertinent data bases (see table 2) revealed two papers by Russian investigator, S.A. Fairushina. Perusal of these papers disclosed that the first dealt with efforts to control the spiny caper, a plant with a very complex root system that is quite destructive of mortar joints. A great many herbicides that readily destroyed the foliage of this plant had little effect on the root system, thus producing little permanent benefit, since it was the root system that damaged the masonry. The second paper had to do with the control of certain flowering plants (Phonerogamae Anthophta) which were destroying the clay brick and plaster brick roofs of architectural monuments in Middle Asia. Both papers dealt with the efficacy of the herbicides; neither mentioned the effect of herbicide on the deteriorating masonry. In both cases, the damage being done by the herb would have made very difficult, if not impossible, any quantitative study of damage done specifically by the herbicide. The only work known to have been done on the effects of herbicides on masonry was accomplished at the Mississippi Test Facility of the National Park Service at Bay St. Louis, Mississippi by Fenn, et al. [13]. Fenn and his co-workers soaked pieces of brick and mortar taken from a deteriorated section of Fort Pickens, Louisiana for one week in 50% aqueous solutions of the isopropylamine salt of N- (phosphonomethyl) glycine (Roundup) and in a 50% aqueous solution of a mixture of 10.2% 4-amino-3, 5, 6-trichloropicolinic acid (trisopropanol amine salt) and 39.6% 2.4-dichlorophenoxy acetic acid (trisopropanol amine salt). This mixture is marketed as Tordon R. The soaked pieces of brick and mortar were dried, then compared with non-soaked samples for resistance to cracking, crumbling, and abrasion. After this experiment, the solutions used for soaking were tested for herbicidal activity. Results are shown in table 3. Fenn and his associates found no change in the masonry samples as a result of the soaking and no change in the herbicidal activity of Roundup due to contact with the masonry.

Table 2. Data Banks

1. Engineering Index
2. Biological Abstracts
3. Chemical Abstracts Chem. Con. 76-86
4. Chemical Abstracts Chem. 707 72-75

Table 3. Germination of brome and radish seeds in water, herbicide solution and herbicide leacheate from brick and mortar.

Treatment	Seed Germination					
	Brome			Radish		
	Total	Germinated	(%)	Total	Germinated	(%)
Water	30	21	70	30	22	77
Roundup (1:9 solution)	20	0	0	30	0	0
Tordon 101 (1:9 solution)	20	14	70	20	10	50
Roundup (brick leacheate)	60	0	0	60	0	0
Tordon 101 (brick leacheate)	60	1	1.7	60	45	75

However they found an appreciable diminution in the activity of Tordon. Since the Roundup did not change in activity discernibly in one week of soaking, it is reasonable to assume that it did not decompose, producing acid decomposition products which might have attacked the masonry. Tordon, however, did show considerable diminution of herbicidal strength indicating that the phenoxy-acetic acid may have decomposed in the manner discussed in Section 5. If so, it is quite possible that the decomposition products may have caused changes in the masonry samples that were too slight to be detected by the procedures used. The findings of Fenn et al. are valuable, but a week is not a sufficient period for such a test and their technique for detecting changes in the masonry was unlikely to have been sufficiently sensitive.

6.2 CRITERIA FOR EVALUATING HERBICIDES

The following are proposed as basic criteria when herbicides must be selected for use on historic structure.

1. The herbicide, when properly applied, should destroy the objectionable plant life.
2. It should not be destructive to masonry or other building components.
3. It should not be destructive of the ecology.
4. It should create a hostile environment for reinfestation by the undesirable vegetation.
5. It should be such that its application would not constitute an extreme hazard to personnel.
6. Its cost should not be exorbitant.

Criteria 1, 2, 3, 4, and 5 concern matters related to plant life and biological effects. Criterion 2, which relates to the subject of this report may be expanded as follows:

a. The herbicide and its decomposition products should not cause defacement or discoloration of the masonry or mortar.

b. It should not reduce the tensile or compressive strength or the impact resistance of the masonry, or in any other way, affect its integrity adversely.

Methods are known for testing for compliance with both divisions of criterion number 4 and for testing for compliance with all of the six criteria listed above.

6.3 OUTLINE OF EXPERIMENTAL PROGRAM FOR STUDYING THE EFFECTS OF HERBICIDES ON MASONRY

For the purpose of selecting desirable herbicides to be used in controlling obnoxious vegetation in the vicinity of masonry which is of historic or artistic importance, a screening program is required. An approach to such a program is suggested in the following paragraph.

Small samples of mortar, concrete, adobe or other masonry material whose stability to herbicides is of interest would be prepared and aged for a reasonable period of time. Three or more samples of each form of masonry to be tested would be exposed to candidate herbicides under conditions which duplicate as nearly as possible the conditions ordinarily observed in the field use of herbicides. Similar samples would be tested under more severe conditions. Additional samples of masonry would be prepared, aged in the same manner but protected from contact with any herbicides.

Microscopic examinations would be made of all samples at regular intervals. After each examination, the exposure to herbicides would be repeated until clearly detectable changes in the masonry samples were observed or until it was determined that no significant change in the masonry sample was occurring. If appreciable changes in any sample occurred early in the course of these experiments, this phenomenon would be studied more thoroughly. Color changes, reflectance, pH measurements or other sensitive tests, would be carried out in efforts to monitor very slight changes very early. The nature of the finding would determine the type of report that would be written to cover this work. Ths approach is infinitely flexible and would be easily modified if conditions or exigencies required such alteration.

7. CONCLUSIONS AND RECOMMENDATIONS

The literature contains very little concerning the effects of herbicides on masonry. The National Park Service has made a beginning in this area. Efforts to control undesirable flora could involve continual use of large amounts of chemicals of various natures. The effects of these chemicals upon masonry must be studied in the absence of the vegetation that they are produced to control if variation in the masonry is to be concomitant. Under actual working conditions, it would be impossible to attribute a given destructive effect specifically to the herb or to the herbicide if both were present. Enough is known about the chemistry of herbicides and their decomposition products to predict that they may have similar effects on the elements of masonry as those demonstrated by related chemicals used for other purposes. In the decomposition reactions, the absence of kinetic data makes impossible the computation of the concentration of decomposition products at any given point; therefore, the effects of these decomposition products must be established by practical experiments if these effects are to be known with certainty. Such experiments would involve exposing the masonry sample to the herbicide in the manner that it would be exposed in actual use for a substantial period (see 6.3). These exposed samples along with controls would then be examined microscopically or by other sensitive techniques which could detect very small changes. From such small changes one may predict with some accuracy, changes which would take place over years, decades or perhaps centuries. This general approach would satisfy the test needs concerning criterion 2 in Chapter 6. Only herbicides meeting the minimum requirements of the other 5 criteria need be tested.

8. ACKNOWLEDGEMENTS

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In preserving historic structures, the control of obnoxious vegetation is a serious problem. To deal with this problem, a number of organic herbicides have been developed by industry. The efficacy of herbicides in the control of plant life has been studied to a great degree; but heretofore, very little has been reported about the possible effects of these chemical plant killers on the materials they are designed to protect. In this work, an exhaustive survey of pertinent literature has been undertaken. Obtaining very little specific information from literature, a correlation has been drawn between the effects of masonry of materials similar in chemistry to herbicides and the effects that would be expected from the herbicides themselves. Methods for checking the validity of conclusions are suggested.

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)*

Herbicide; masonry; historic structures; degradation; acidic, alkaline

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