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Salt Weathering: A Selective Review

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Abstract: The past decade has seen a growing scientific interest in the still poorly understood subject of salt weathering, a phenomenon with significant cultural and economic consequences. This interest has led to an increase in research results and growing clarification of the roles salts play in weathering and decay. There are now over 1,800 research articles on the topic of salt weathering originating from several disciplines, as well as over 6,000 references on the general problems of building material decay. In order to navigate such a vast collection of data and knowledge, this article describes the multidisciplinary nature of the study of salt damage to porous building materials, provides a framework for considering the complexity of salt damage, and serves as a selective literature survey largely focused on recent work and those articles with relevance for conservation.

> "In time, and with water, everything changes"
>
> —Leonardo da Vinci

Over the past ten years, there has been increasing scientific interest in building material decay phenomena, especially the incompletely understood subject of salt weathering. The reaction of salts with moisture in rock outcrops produces an intriguing array of weathering forms such as tafoni, honeycombs and pedestal rocks, as well as abundant rock debris (Goudie & Day 1980; Mustoe 1982; Smith 1994). Salt weathering of building materials can be equally destructive (Haynes, O'Neill *et al.* 1996). Salt weathering is widely recognized as one of the primary agents in the deterioration of historical architecture, structures in archaeological sites, and archaeological objects (Schaffer 1932; Lewin 1982). Understanding and finding ways to reduce damage caused by salts holds great significance for the conservation of material cultural heritage (Torraca 1982; Amoroso & Fassina 1983; Goudie & Viles 1997). Moreover, damage to modern concrete building foundations caused by salts has recently been the subject of litigation leading to multi-million dollar settlements and judgments (Haynes 2002; Kasdan, Simonds *et al.* 2002). Salt weathering is indisputably a process with profound cultural and economic consequences.

Salts have long been known to damage porous materials (Herodotus 420 B.C.; Luquer 1895) through the production of physical stress resulting from the crystallization of salts in pores (Taber 1916; Jutson 1918). Salts can damage stone and other building materials through a range of other mechanisms as well, such as differential thermal expansion, osmotic swelling of clays, hydration pressure, and enhanced wet/dry cycling caused by deliquescent salts (Smith 1994; Nash 2000). A 1997 book by Goudie and Viles summarizing salt weathering hazards raised awareness of the problem and broadened the field of investigation in recent years. Despite this, many unresolved issues related to salt weathering remain, ranging from the details of the damage mechanisms (Flatt 2002) to the interactions between substrate, environment and salt type (Charola 2000; Blaeuer *et al.* 2001), and the expression of this process in the landscape (Smith, Warke *et al.*

2000) and in building stone (Kuchitsu, Ishizaki *et al.* 2000; Schwarz, Gervais *et al.* 2000).

This article is intended to describe the multidisciplinary nature of the study of salt damage to porous building materials; communicate the complexity of salt weathering and discuss open questions; and serve as a limited literature survey largely focused on recent work and those articles with relevance for conservation.

Background

The majority of the published literature on salt weathering originates from a collection of distinct disciplines, each responsible for specific contributions: geomorphology, environmental science, geotechnical and materials science, geochemistry, and, lastly, conservation, perhaps the most multi-disciplinary of the fields. There is a rich literature in geomorphology on salt weathering phenomena, particularly that published by scientists from the United Kingdom (Smith 1994; Goudie & Viles 1997). Environmental scientists have studied the important role of salts in atmospheric aerosols and their fallout (Pósfai, Anderson *et al.* 1995; Torfs & Van Grieken 1997). The geotechnical field has dealt with expansive sulfate soils, or salt heave, (Xiaozu, Jiacheng *et al.* 1999), concrete decay from salt crystallization (Haynes, O'Neill *et al.* 1996; Mehta 2000; Haynes 2002), and testing of building materials using standardized salt crystallization tests such as ASTM aggregate sound test C88-90 (ASTM 1997), and RILEM PEM/25 (RILEM 1980). Materials scientists have provided useful insights on the fundamental mechanisms of salt attack (McMahon, Sandberg *et al.* 1992; Scherer 2000). In the geochemical literature, the concepts of pressure solution, force of crystallization, and displacive growth are important to understanding that salt attack is a part of a larger set of interrelated behaviours (Carstens 1986; Maliva & Siever 1988; Minguez & Elorza 1994). Finally, the conservation literature is vast and especially rich in case studies and documentation of methods of mitigating salt damage (Schwarz & Roesch 1993; Young 1995; Siedel 1996; Charola, Henriques *et al.* 1998; Price 2000; Unruh 2001). German speaking conservation scientists, conservators and architects have published a number of important books on the topic of salts and cultural heritage (Hammer and Tinzl 1996; Arendt and Seele 2000; Kraus 2002).

Nevertheless, much of the conservation literature on salts can be difficult to survey, since many articles are published in conference proceedings (Price 1996). Two recent summaries of the literature illustrate the breadth of applicable research: an overview of the role of salts in the deterioration of porous materials by Charola (2000) and a discussion of salts and crusts by Steiger (2002).

Following these recent efforts, this paper presents a current survey of the multidisciplinary contributions to the field of salts and building deterioration. A large bibliography of research articles on salts and building deterioration phenomena was assembled in order to facilitate the integration of these related researches. Some observations based on this survey are discussed below.

A recitation of the sometimes significant, sometimes subtle, differences in terminology and definitions employed in the literature of salt weathering reveals the multidisciplinary nature of the scientific literature. The same basic phenomena are variously described in different disciplines and countries as salt attack, salt damage, salt crystallization, salt crystallisation (UK), haloclastisme (French), and salzsprengung (German). Less common terms include salt burst, salt exudation, haloclasty, salt decay, salt crystallization pressure, crystal wedging, salt fretting, and salt heave in soils.

In the geomorphology literature, the terms "honeycomb weathering", "alveolar weathering", and the less common descriptors "stone lattice", "stone lace", and "fretting" (Mustoe, 1982), reference a characteristic form of salt weathering in stone defined by irregular cavities in otherwise regular rock surfaces, although in recent publications those terms are increasingly discarded in favor of the term "tafoni" for cavities large and small. Substantial research has been performed by geomorphologists on tafoni (singular: tafone), a term that normally refers to pits and caverns in rock faces resulting from extensive salt weathering of stone in salt-rich and desert environments (Kirchner 1996; Turkington 1998). Tafoni also occur in humid temperate environments; however, it is not yet clear that salt weathering plays a role in the formation of tafoni in this type of climate (Mikulas 2001).

In the conservation literature, the term "rising damp" is widely used to characterize salt weathering, but in Australia this process of moisture and salts mobilized by capillarity and deliquescence is regionally referred to as "salt damp" (Young 1995). Many buildings and sites are also affected by salt-rich aerosols, which may accumulate and be mobilized by "falling damp" or "penetrating damp."

In the concrete literature, the terms "salt attack" and "sulfate attack" are sometimes incorrectly used interchangeably (Mehta 2000), and recent authors have proposed the terms physical salt attack or salt hydration distress as distinct from the chemical reaction of salts with concrete (Haynes, O'Neill *et al.* 1996; Hime, Martinek *et al*. 2001; Haynes 2002). Stone conservators typically use the terms desalting, salt extraction, or desalination, referring to salt reduction methods using immersion, intermittent washing, poulticing or compress treatments.

Given the variety of terms used and the diversity of disciplines researching the topic of salt weathering, it seems clear that at least some of the apparent contradiction in the literature is merely the result of differences in terminology (Hime, Martinek *et al.* 2001).

Salt Damage: Dealing with Complexity

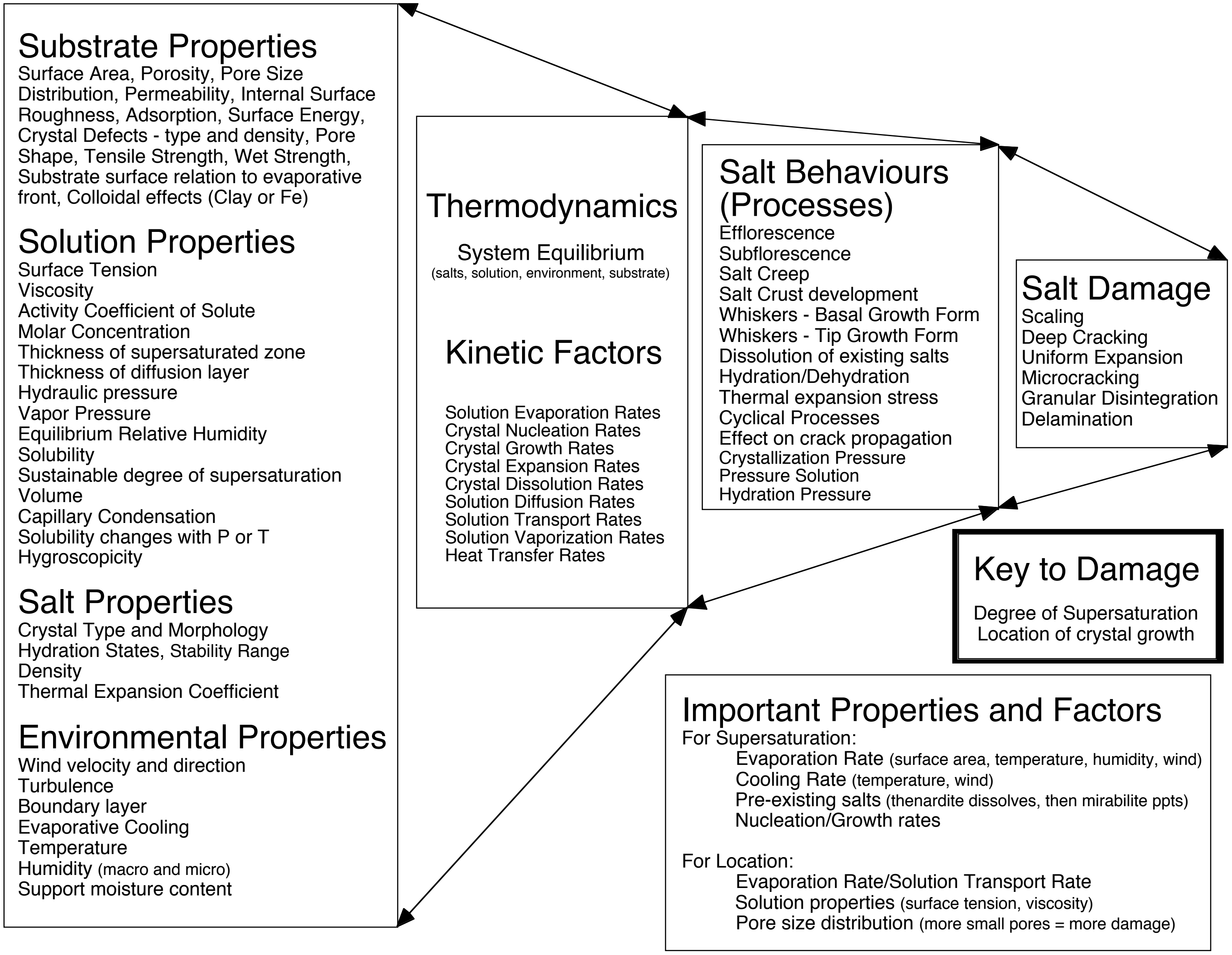
Two major difficulties encountered by those investigating salt weathering are the multiple variables involved and the fact that the physiochemical reactions of greatest importance occur along thin, nanometer-scale films inside a porous solid. Such interactions are inherently difficult to study *in situ* and *in tempo*, further inhibiting understanding of the degree of interaction between possible variables. To aid understanding of the variables, it is useful to classify them as properties of the substrate, solution, salt type, or environment (Fig. 1). The properties interact in specific ways determined by thermodynamic equilibria and kinetic factors to produce a range of salt behaviours.

It is important to note that not all salt behaviours result in deterioration. For example, the production of surface efflorescence is often impressive and highly visible, but generally results in little damage. Likewise, salt creep can move salts over the surface of materials but, as the name implies, is a surficial process. Salts such as magnesium sulfate can actually bind together (i.e., cement) previously fractured material.

Understanding the properties that contribute to a particular weathering effect, and differentiating between benign and damaging salt behaviors, allows the investigator to focus on the latter. Typical damage behaviors or processes for salts can include surface scaling, deep cracking, expansion, granular disintegration, surface powdering and microcracking. For particular damage mechanisms, such as crystallization pressure, the degree of damage (sometimes measured as the damage factor, damage function or dose-response function) is often attributable to specific properties and can be the single most important metric (Viles 1997). For example, in the case of sodium sulfate crystallization, the degree of supersaturation and the location of crystallization appear to be the keys to understanding the degree of damage (Rodriguez-Navarro & Doehne 1999a). In turn, there are a few important properties and kinetic factors, such as the evaporation rate, that largely control how much damage results from this mechanism (Fig. 1).

By focusing on the nature (i.e., moment, location and type) and, especially, the degree of salt damage, the field of variables, or properties, directly responsible can be narrowed considerably and investigators can begin to answer the questions most relevant to specific damage types. Why are certain types of stone much more vulnerable than other types to salt damage (Goudie 1999b)? Why are certain salts much more damaging than other salts (Hoffmann & Grassegger 1995; Aires-Barros & Mauricio 1997; Rodriguez-Navarro & Doehne 1999a)? Is damage caused mostly by relatively rare environmental events (rapid cooling) or cumulative everyday stresses (humidity cycling)? What are the long-term effects of various conservation treatments, such as desalination or consolidation, on salt damage? How can desalination and preventive conservation efforts be enhanced? Can general agreement be achieved regarding the fundamental mechanisms of salt weathering? What are the appropriate equations to use in calculating crystallization pressure (Duttlinger & Knöfel 1993; La Iglesia, Gonzalez *et al.* 1997;

Figure 1. Diagram of properties, factors and behaviours in the salt crystallization process.



Substrate Properties

Surface Area, Porosity, Pore Size Distribution, Permeability, Internal Surface Roughness, Adsorption, Surface Energy, Crystal Defects - type and density, Pore Shape, Tensile Strength, Wet Strength, Substrate surface relation to evaporative front, Colloidal effects (Clay or Fe)

Solution Properties

Surface Tension
Viscosity
Activity Coefficient of Solute
Molar Concentration
Thickness of supersaturated zone
Thickness of diffusion layer
Hydraulic pressure
Vapor Pressure
Equilibrium Relative Humidity
Solubility
Sustainable degree of supersaturation
Volume
Capillary Condensation
Solubility changes with P or T
Hygroscopicity

Salt Properties

Crystal Type and Morphology
Hydration States, Stability Range
Density
Thermal Expansion Coefficient

Environmental Properties

Wind velocity and direction
Turbulence
Boundary layer
Evaporative Cooling
Temperature
Humidity (macro and micro)
Support moisture content

Thermodynamics

System Equilibrium
(salts, solution, environment, substrate)

Kinetic Factors

Solution Evaporation Rates
Crystal Nucleation Rates
Crystal Growth Rates
Crystal Expansion Rates
Crystal Dissolution Rates
Solution Diffusion Rates
Solution Transport Rates
Solution Vaporization Rates
Heat Transfer Rates

Salt Behaviours (Processes)

Efflorescence
Subflorescence
Salt Creep
Salt Crust development
Whiskers - Basal Growth Form
Whiskers - Tip Growth Form
Dissolution of existing salts
Hydration/Dehydration
Thermal expansion stress
Cyclical Processes
Effect on crack propagation
Crystallization Pressure
Pressure Solution
Hydration Pressure

Salt Damage

Scaling
Deep Cracking
Uniform Expansion
Microcracking
Granular Disintegration
Delamination

Key to Damage

Degree of Supersaturation
Location of crystal growth

Important Properties and Factors

For Supersaturation:
- Evaporation Rate (surface area, temperature, humidity, wind)
- Cooling Rate (temperature, wind)
- Pre-existing salts (thenardite dissolves, then mirabilite ppts)
- Nucleation/Growth rates

For Location:
- Evaporation Rate/Solution Transport Rate
- Solution properties (surface tension, viscosity)
- Pore size distribution (more small pores = more damage)

Benavente, Garcia del Cura *et al*. 1999; Scherer 1999; Charola 2000)? Can the large number of laboratory simulation studies of salt weathering be reconciled with field data and explained using existing theories? Can salt weathering forms such as tafoni be accurately modeled using existing knowledge? How do the behaviors of water, salts, and substrate compare at the important mesopore scale (2-50 nanometers) versus the macro scale (Brown 2001; Hochella 2002)? These complex questions, together with such basic questions as how the hydration of salts progresses and how crystallization pressures are sustained *in situ*, are still in need of study (Charola & Weber 1992; Doehne 1994; Steiger, Beyer *et al*. 2000). Nevertheless, the research performed in recent years and surveyed herein has significantly advanced overall understanding of the damage caused by salt weathering processes.

Deterioration due to salts: Theory and Laboratory Observations

Several important advances in salt damage arising from theoretical and laboratory research have application for the conservation field and deserve highlighting. The research summarized here addresses the precise nature of the relationship between salt stress and material pore size; how supersaturated solutions develop and are maintained; the contradictory behavior of the common salts halite and gypsum in the field as opposed to the laboratory; and the important role of humidity cycling in salt crystallization.

George Scherer (1999; 2000) has recently advanced the theoretical framework for understanding stress caused by the crystallization of salts in pores. This research helps explain why salt crystallization may not damage certain materials: in some cases, the local stress field is not large enough to propagate critical flaws in the material. While stones with large pores tend to fare better than those with abundant small pores, Scherer's work points out that large pores effectively behave like small pores when filled with salt. Scherer (1999) finds that the maximum pressure salt crystallization can achieve is highly dependent on pore size, predicting that most of the damage occurs when salt growth migrates from larger to smaller pores in the size range of 4 nm to 50 nm. Scherer's work clarifies the visual model of how salts growing on the nanometer-scale thin film between salt and stone can result in stresses that exceed the strength of most porous building materials. Putnis & Mauthe (2001) have noted that halite cementation in porous sandstone reinforces the observation that larger pores fill before smaller pores, confirming that fluids in small pores can better maintain higher supersaturation.

It has long been known that the solution must be supersaturated, in order for salt crystallization to damage porous materials (Goudie & Viles 1997). What has been unclear until recently is how, in a material with abundant nucleation sites, such a supersaturated solution was created and maintained.

Dei et al. (1999) investigated the experimental crystallization of potassium nitrate salt in Italian marble, sandstone and travertine samples. They found that the pore size distribution was shifted towards larger pores and the porosity was reduced after salt crystallization. This appears to be the result of blocking of micropores due to crystallization of salts in the zone between large and small pores. Thermal analysis of the samples to identify the salts showed that the shape and timing of the exothermic peak indicative of crystallization varied depending on the pore system and stone type.

The important relationships between the rate at which supersaturation increases, nucleation, and pore size distribution was studied by Putnis et al. (1995). They found that nucleation was suppressed in fine-grained substrates in experiments with gypsum and other salts. This work provides provides another explanation for empirical observations that stones with abundant micropores are often have a higher decay rate in standard salt crystallization tests. Also, it helps explain how high supersaturations may be obtained in materials with a high internal surface area.

Kashchiev and Van Rosmalen (1995) present a way to calculate the supersaturation a solution can maintain in a pore of a given radius before crystallisation occurs. Due to the Laplace effect of curvature, in small pores (less than 1 micron) a higher supersaturation can be reached before crystallization begins. Pores also alter the crystallization of ice, dramatically reducing the normal freezing point (Baker et al. 1997).

Some authors have taken the idea of crystallization pressure and applied it to less soluble minerals such as calcite (McBride and Picard 2000), suggesting that crystallization of calcite in a calcite-rich rock may result in the production of tafoni. However, conventional salt crystallization theory requires the pore wall and the salt to be incompatible in order to generate crystallization pressure (Scherer 2000), so some other mechanism may be at work.

Crystallization can be characterized as needing a driving force, such as a supersaturated solution, to create and sustain it. The generally accepted kinetic pathways to produce such a solution or mode of supersaturation generation are rapid cooling, for salts with a strong solubility dependence on temperature, and rapid evaporation for the remainder of salts (Rodriguez-Navarro, Doehne *et al.* 1999). A third reaction pathway was first noted by Chatterji & Jensen (1989), more recently by Doehne, Selwitz *et al.* (2001), and in detail by Flatt (2002). This pathway is the presence of fine-grained pre-existing salts that dissolve rapidly to produce a supersaturated solution, such as is caused by the wetting of thenardite, leading to the rapid precipitation of mirabilite from solution, as observed in the environmental SEM (Doehne 1994; Rodriguez-Navarro & Doehne 1999a).

Another unresolved conundrum is the contrast between severity of damage in the field caused by halite and gypsum (Kirchner 1996), two of the most common salts, as compared to their relatively benign behavior in the laboratory (Smith & McGreevy 1988; Goudie & Viles 1997; Robinson & Williams 2000). Understanding gypsum's behaviour is especially important to stone conservation in view of the common occurrence in the field of "black crust" containing abundant gypsum, a product of the reaction of regional sulfate air pollution with building stone (Verges-Belmin 1994; Wittenburg & Dannecker 1994). Despite the ongoing reduction of sulfate air pollution over the past 20 years, the rate of loss of carbonate stone surface has not slowed proportionally at sites such as St. Paul's Cathedral in London. This discrepancy has been attributed to salt crystallization damage from pre-existing gypsum (Trudgill, Viles *et al.* 1991).

Gypsum also plays a significant role in the typical problems of masonry buildings near the sea. Salt weathering related to sodium chloride and gypsum from sea spray salts and soil capillary waters is addressed in a recent case study documenting the decay history of the site of Civil Palaces in Alicante, Spain (Louis, Del Cura *et al.* 2001). The desalination of porous building materials typically results in the non-proportional removal of salts (Bromblet & Verges-Belmin 1996). If gypsum is present, the removal of hygroscopic salts may decrease its mobility since the solubility of gypsum is significantly increased by the presence of sodium chloride and other salts (Robinson & Williams 2000).

Other open questions pertaining to salt damage of porous materials are the role of the range of humidity cycling, the potential presence of damage thresholds, and the degree to which damage over the long term can be reduced by decreasing humidity fluctuations. Experiments on ceramic tiles loaded with NaCl, Na_2SO_4 and $CaSO_4.2H_2O$ and simulating the humidity cycles (43%-55% RH and 0%-11% RH) of museum display cases (Nunberg & Charola 2001) over the course of six months found that measurable deterioration of the tiles had taken place. This ran contrary to expectations that no damage would occur in a closed environment with limited humidity cycling well below the deliquescence point of the salt (equilibrium relative humidity or RHeq). The discovery of damage suggests that while environmental control significantly reduces the rate of damage, additional research is needed to understand the limits of environmental control and how the damage was produced. In this example, the mode of solution supersaturation is not clear. However, the filling of larger pores with salt and the creation of smaller pores may play a role, as might capillary condensation of water in fine pores combined with limited humidity cycling exerted over time (Rucker, Holm *et al.* 2000). The results of Nunberg and Charola (2001) suggests that environmental control may provide less protection than previously believed (Price 2000).

The standard salt crystallization tests (ASTM and RILEM) for building materials, where sample blocks are repeatedly immersed in saturated solutions of sodium sulfate and then dried, are widely used to estimate resistance to frost damage and general durability (Price 1978; Marschner 1979). Nevertheless, there has been some controversy over the applicability of the tests, given the severity of the test and the sometimes inconsistent results (Sheftick 1989; Hunt 1994). Recent research has addressed this problem from two directions: examining the way the test actually operates (Rodriguez-Navarro, Doehne *et al.* 2000a) and proposing new standard test methods which more closely simulate typical conditions (Benavente, Ordóñez *et al.* 2001). The assumption of many researchers has been that the damage from this test occurred due to the hydration of sodium sulfate. More recent work suggests that direct crystallization of thenardite (Na_2SO_4) may occur (Rodriguez-Navarro, Doehne *et al.* 2000a) in addition to rapid mirabilite ($Na_2SO_4.10H_2O$) crystallization during immersion (Doehne, Selwitz *et al.* 2001; Flatt 2002). Goudie (1999a) found that salt crystallization tests of limestone were poor predictors of frost resistance. Efforts to discover parameters that correlate with building stone durability continue, with emphasis on those that can be measured more easily than salt crystallization resistance (Moh'd, Howarth *et al.* 1996; Ordonez, Fort *et al.* 1997; Nicholson 2001).

New Methods for Studying Salt Weathering

An important turning point in the advancement of salt weathering studies has been the development of methods that provide data on material behavior *in situ* and *in tempo*, such as ultrasonic field testing (Goudie 1999b; Simon & Lind 1999), acoustic emission (Storch & Tur 1991; Grossi, Esbert *et al.* 1997), time-lapse and Environmental Scanning Electron Microscopy (ESEM) (Doehne 1997; Rodriguez-Navarro & Doehne 1999b), and three-dimensional imaging methods such as NMR (Rucker, Holm *et al.* 2000) and micro-CT scanning (Degryse, Geet *et al.* 2001). Such methods add the third and fourth (time) dimensions to previous studies and aid in separating the effects of salts from other decay phenomena. For example, Pel (2000) found that the absorption of a concentrated NaCl solution in calcium-silicate brick, as seen when using NMR, revealed a sharp wetting front. Unexpectedly, the sodium was clearly seen to lag behind the moisture profile. Little sodium, if any, was observed near the wetting front, an apparent result of the interaction of sodium ions with the pore surface. In tests of 21 limestones using non-destructive methods, Goudie (1999b) noted that even durable stones which showed no visible decay after salt crystallization tests were found to have suffered significant decreases in their modulus of elasticity values, indicating loss of strength. Limestone durability was generally found to correlate with high values of modulus of elasticity, lower water absorption capacities, high densities and low salt uptakes.

Deterioration due to Salts: Field Observations

A wide range of important field observations of decay to porous building materials over several decades has been summarized by Arnold and colleagues (Arnold & Küng 1985; Arnold & Zehnder 1985; Arnold & Zehnder 1988; Arnold & Zehnder 1991). These observations clearly establish the strong relationship between the rate of salt decay and the environmental conditions. Arnold discusses the morphology of the salt crystals and its relationship to the support moisture content. With a relatively dry substrate with a slow evaporation rate, the crystals take on an acicular form; when more moisture is present, a salt crust may form (Arnold & Küng 1985; Arnold & Zehnder 1985). Subsequent observations of climate and salt behavior in six churches over the course of five years showed that… "hygroscopic salts crystallize periodically according to the variations of relative humidity and to a much lesser extent of temperature. Within continuously heated rooms crystallization and decay are related to variations in relative humidity caused primarily by the heating and subordinately by natural variations of the outside climate. In non heated rooms seasonal variations of temperature also induce periodic crystallization." (Arnold & Zehnder 1988). Arnold also documented the fractionated pattern of salts in walls with rising damp, dividing the wall into four zones in vertical succession differentiated by degrees of damage and salt content. The lowest zone was perpetually damp with salts remaining in solution, succeeded by the zone of greatest damage and, finally, the uppermost zone marking the limit of rising moisture caused by deliquescent salts. The highest salt content does not always correspond to the greatest damage, since ion mixtures that are strongly hygroscopic will not crystallize under normal conditions. The sources of the salts are attributed by Arnold to the extensive use of cement-based mortars and concrete in large restoration programs over the past 50 years, as well as salts from air pollution and rising damp. Soluble salts from cement-based mortars and concrete are still today an important cause of decay to historic building materials, despite efforts to improve cement quality and reduce the overall use of these materials (Moropoulou 2000).

A brief review of other more recent field observations on salt weathering is considered next. Some useful insights into rates of salt weathering have recently been gained through the measurement of tafoni depth using rock surfaces with a range of known ages (Matsukura & Matsuoka 1991; Sunamura 1996; Norwick & Dexter 2002). Tafoni are cavities or pits in otherwise homogeneous rock surfaces. The current consensus appears to confirm the salt weathering origin of most tafoni (Pye & Mottershead 1995; Mikulas 2001; Norwick & Dexter 2002). For tafoni that has developed over the last century, linear recession rates are estimated at 0.6 to 5.2 mm/year (Sunamura 1996). Tafoni depth was also found to develop twice as fast as the width of the pit, providing additional confirmation for the salt weathering origins of tafoni. More recent analysis has shown that while there is significant scatter in the depth measurements, it appears that the weathering rate is actually non-linear, with a time lag followed by rapid recession and then a much slower rate of loss as the tafoni age. This pattern generates a sigmoidal or “S” curve, a typical example of which is given by the following equation:

$$D = b1 + \mathrm{e}^{(b2+(b3/t))} \qquad (1)$$

where D is the depth of the tafoni, t is the age of the surface and $b1$, $b2$, and $b3$ are constants solved for mathematically (Norwick & Dexter 2002).

A speculative explanation for the non-linearity suggests that the rate of loss is first limited by low salt concentrations, then becomes dominated by a positive feedback loop with an increasing rate of damage related to increasing salt concentration, followed by a negative feedback loop as the scale of the tafoni reaches a point where it apparently no longer allows efficient salt weathering (Norwick & Dexter 2002). This non-linear pattern is typical for many other natural weathering processes (Goudie 1995). Such non-linearity may help explain why salt weathering rates measured in laboratory settings have been much more rapid than those typically found in field studies. While erosion rate data from the study of stone structures for conservation is surprising uncommon, such patterns of low rates of decay, followed by a rapid increase have been noted by Charola (2000) and Snethlage, Wendler *et al.* (1996). The fact that field salt weathering rates are typically non-linear and may show a low initial rate of loss suggests that stone conservators should interpret short-term observations of overall stability of vulnerable structures, such as wall paintings, with caution.

One of the advantages marble possesses for resisting salt damage is its minimal porosity and tendency not to have a strong capillary suction (which can lead to rising damp). However, recent studies of the marble monuments at the archaeological site of Delos, Greece have shown that salts deposited from atmospheric aerosols are responsible for damage of marble sculpture (Jeannette 2000; Chabas & Jeannette 2001). Additionally, recent work on soil chemistry in Hawaii using strontium isotope data has found that about 50% of the calcium in soil samples from a coastal location originated from salt spray, not soil mineral weathering as expected (Whipkey, Stewart *et al.* 2000). This suggests that salt spray has a strong effect on the local environment, perhaps dependent on overall wind and storm patterns.

An extensive survey and analysis of bedrock in the Valley of the Kings, Luxor, Egypt recently resulted in warnings concerning the hazards to ancient tombs from swelling clays and salts activated by moisture from the exhalations of tourists as well as flash floods (Wüst & McLane 2000; Wüst & Schlüchter 2000). Expansion from hydration of anhydrite and crystallization cycling of abundant sodium chloride where cited as important decay mechanisms in important Royal tomb sites, such as Seti I, which have seen a rapid increase in stone loss since their opening by archaeologists.

Warke and Smith (2000) suggest that "Salt weathering is a threshold phenomenon where decay is manifest only when a stress/strength threshold is crossed." This conclusion is based on the analysis of two similar sandstone blocks, one sound and one damaged, with similar salt content and distribution. They found that NaCl was deeper and more mobile in the stone (6 cm) than expected from surface damage (2 cm). A strong correlation of decay rate with environmental variation has recently been documented by Cantón, Pini *et al.* (2001). In a range of experiments that the weathering rate of a calcareous mudstone was found to most closely follow the number of wetting/drying cycles. Some useful cautionary notes on the real world distribution of salts in sandstone can be found in (Turkington & Smith 2000). The researchers concluded that "The widely held perception that urban environments are 'dry' with shallow surface wetting of building stone does not appear to hold true for certain building stone."

Salt weathering is traditionally associated with coastal and desert environments (Theoulakis & Moropoulou 1999), however regions with a tropical monsoon climate may also be effected by salt weathering due to the strong contrast between the rainy and dry season. Two recent examples are the famous temples at Angkor in Cambodia (Uchida, Nakagawa *et al.* 2000) and the brick monuments in Ayutthaya, Thailand (Kuchitsu, Ishizaki *et al.* 2000). At Angkor, one of the main causes of deterioration was identified as crystallization pressure from gypsum and phosphate minerals growing between exfoliated layers of sandstone. The sulfur and phosphate are derived from rain water leaching of bat guano, which subsequently moves into the stone through rising damp (Uchida, Nakagawa *et al.* 2000). At Ayutthaya, gypsum efflorescence is found in the rainy season, with more soluble salts such as thenardite appearing in the dry season. Most damage occurs at the beginning of the dry season to brick surfaces, and Kuchitsu Ishizaki *et al.* (2000) propose methods to reduce water impregnation of the bricks in the rainy season as a conservation measure.

Interrelationship between Salt Weathering and Other Decay Mechanisms

With an understanding of the properties necessary for a particular salt damage mechanism to be activated, it is possible to contemplate modifying those properties to better understand the damage mechanism and to develop preventive conservation measures. Changing a parameter such as surface tension using surface-active agents was recently found to have a strong effect on damage from sodium sulfate crystallization since it directly affects where salts crystallize (Rodriguez-Navarro, Doehne *et al.* 2000b). The authors speculated that biologically produced surfactants might have a similar effect. Two articles have subsequently found that the presence of bacteria, associated biofilms, and biopolymers greatly increases the damage caused by salts (May, Papida *et al.* 2000; Papida, Murphy *et al.* 2001).

The relationship between salts and frost weathering was explored recently by Williams & Robinson (2001). The authors extended the range of salts known to intensify frost weathering (potassium and ammonium alums) and confirmed the significance of halite in the process. Williams & Robinson (2001) show that the degree of damage to stone varied greatly depending on the combinations of salts involved. While most salts contributed to damage proportionally when used in combination, some salt mixtures caused intensified damage out of proportion to the component salts. The topic of salt mixtures and their effect on salt damage and other decay mechanisms is complex since the behaviour of the mixture cannot be predicted from single salt data (Steiger & Dannecker 1995; Steiger 1996; Steiger & Zeunert 1996). However, the use of computer models such as the semi-empirical ion interaction model of Pitzer provides an important predictive tool for studying most field situations where salt mixtures are the rule (Clegg & Brimblecombe 2000; Steiger, Beyer *et al.* 2000).

Dragovich (1997) studied the weathering of marble tombstones near Sydney, Australia and was surprised to measure a low weathering rate (540 microns/ 100 years). The seaward facing sides of the marble blocks, which were exposed to greater salt input, unexpectedly weathered more slowly than the landward side. This suggests that even in coastal environments salt weathering is only one aspect of the full weathering process.

Prevention, Mitigation, and Treatment Options

Avoiding damp is a fundamental principle for maintaining any structure where porous materials are used (Ashurst & Ashurst 1988). The most important mitigation method is prevention, through the physical separation of building materials from soil moisture and salts with the traditional "damp-proof course." This is typically an impermeable barrier such as plastic (current structures), glazed brick, bitumen, or other material. The service life of the plastic sheeting used in modern concrete slab construction is not well known. In existing salt-laden structures, salts can be removed through poulticing and in some cases the affected masonry may be replaced. However, in the past few decades, the cost of such replacement has led to increasing use of chemical "damp-proof courses" in the form of injected siloxane. Below, a range of treatment options and research into the reduction of salt damage is discussed.

Salts tend to cause damage in building materials when activated by a change in humidity, temperature or the presence of liquid water. If the composition of the salts is known, then calculating the stability range (RH and temperature) for the salts allows establishment of the appropriate conditions for minimizing damage due to a change of state. Establishing such controlled conditions should substantially extend the life of the material. An important new report by Price and colleagues (2000) documents the development of an expert system to provide such information and is an extremely useful repository of detailed thermodynamic and kinetic information concerning individual salts, salt mixtures, and their calculated and actual behavior. Other researchers have also explored this approach (Aires-Barros & Mauricio 1997). Unfortunately, controlled environments for stone objects and structures tend to be the exception rather than the rule, and field experience suggests that damage tends to occur whenever significant concentrations of salts are present in porous masonry (Andreas Arnold, personal communication).

The practice of poulticing salts using highly absorbent materials such as clays or wood pulp has long been employed by stone conservators to remove salts from important wall paintings and carved stone. Study of the effectiveness of these methods has shown that, while chloride and nitrate are readily removed, sulfate removal is more difficult. Artificial anionic clays (hydrotalcites) are recommended for enhanced extraction of the less soluble gypsum (Vicente & Vicente-Tavera 2001). Vicente & Vicente-Tavera (2001) also noted the loss of extraction efficiency under conditions of high humidity. Simon, Herm *et al.* (1996) found that a new high porosity compress render was generally more effective at removing salts than a traditional sacrificial lime plaster, although the introduction of water to the surface during the treatment increased nitrate concentrations higher up on the wall. Siedel also noted the difficulty of controlling desalination (Siedel 1996), achieving results that were "contradictory and sometimes even undesirable on large monuments."

Two desalination methods (total immersion and intermittent washings) were studied recently for their potential for use in the removal of salts from ceramic tiles by Freedland & Charola (2001). The authors found that for more soluble salts, such as NaCl, repeated washings were more effective, while long-term immersion was more efficient for less soluble salts such as gypsum as well as for sodium sulfate in ceramic tiles with fine pores. Based on a substantial body of experimental work, Charola, Freedland *et al.* (2001) have developed a reproducible method to calculate the salt remaining in an object after desalination.

An interesting effort to move sodium chloride from a brick vault into an adsorbing mortar above the vault used differential humidity to mobilize the salts (Larsen & Bøllingtoft 1999; Larsen 2001). With a low relative humidity above the vault and a moist environment beneath (set below the deliquescence point of the salts), it was found that the kinetics for NaCl movement solely attributable to humidity are slow, and that NaCl most readily moves into the mortar (from damaged bricks) when liquid water is present.

A newly proposed approach mitigating salt weathering is the reduction of the interfacial energy between the salt and the stone using a specially selected non-swelling polymer with the appropriate surface properties (Scherer, Flatt *et al.* 2001). By placing such a barrier between the pore walls and salts, it is anticipated that greater surface energy compatibility may be achieved and the salts should stop growing when they come in contact with the polymer.

Long experience with salt weathering of building foundations in Adelaide, Australia, a region with a Mediterranean climate and salt-rich soil, has resulted in a host of practical methods for mitigating this damage (Blackburn & Hutton 1980; Young 1995). Young found that decay is most commonly found at the base of walls, where solutions migrating from local salt-rich soils damage masonry, mostly in cases where a damp-proof course is either lacking, has been damaged or was bridged by later masonry. The most effective treatment is replacement of the salt-laden stone and reinstallation of a damp-proof course. The high cost of this treatment is limits its use to severe cases or situations where sufficient funding is readily available. The most common treatment is injection of waterproofing agents (siloxane) to provide a chemical damp-proof course and desalination of the salt-laden masonry. The longevity of these treatments remains uncertain, with some failures attributed to problems with desalination or incomplete injection. Saline soils are an increasing regional problem in south and southeast Australia, and salt weathering of heritage structures in this region has begun to mount (Spennemann 2001).

Planetary Salt Weathering

Salt weathering is a global phenomenon, present in Antarctica as well as Death Valley. This diversity has been appreciated by geologists and planetary scientists, who have suggested that salt-weathering mechanisms may help explain unusual features on Mars (Malin 1974). Photographs from Mars taken at the Pathfinder landing site reveal features remarkable similar to honeycomb weathering structures on Earth (Rodriguez-Navarro 1998). McCord (2001) found that under conditions typically found on the Jovian moon Europa, magnesium sulfate could remain hydrated on a geologic time scale.

Summary and Future Work

As of this writing, there are over 1,800 references in the scientific literature on the topic of salt weathering of porous materials and over 6,000 references addressing general problems of building material decay. Many of these articles describe unique case studies that may be difficult to interpret or apply to other situations. In order to navigate this vast collection of data and knowledge borne of disparate disciplines, this article serves as an organizing framework to consider the complexity of salt damage, presents an overview of multidisciplinary research and open questions on this topic, and provides a limited literature survey largely focused on recent work and those articles with relevance for conservation. The research surveyed here has clearly advanced our understanding of the damage caused by salt weathering processes, and raises important questions for the conservation of material heritage. Future investigation into salt weathering should include more quantitative measurements of field weathering rates and laboratory studies using methods such as three-dimensional imaging to further illuminate the relationships between salt weathering mechanisms and the properties of solution, substrate and environment, and to contribute to a better understanding of the action and kinetics of salt mixtures in weathering stone. Finally, improving methods to prevent and mitigate salt weathering of building materials clearly deserves more effort and attention.

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