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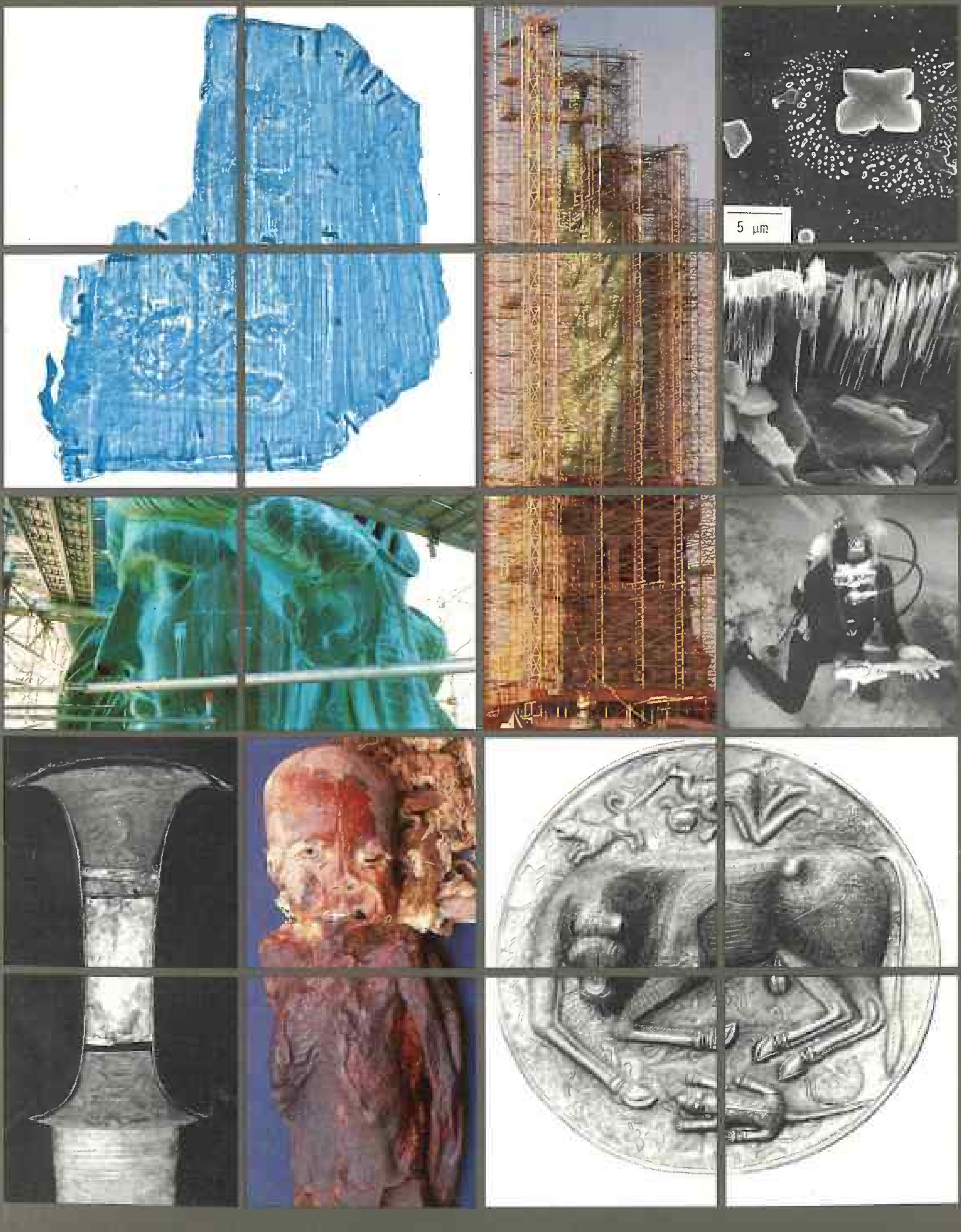

The Deterioration of Ceramics by Soluble Salts and Methods for Monitoring their Removal

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Abstract : *The sources of soluble salt contaminants in archaeological ceramics are examined during burial and after excavations. The behavoiur of the hydrous and anhydrous salts and the physical and chemical processes of deterioration are described. Three conservation methods for the treatment of contaminated ceramics are considered. Two methods, the control of the ambient relative humidity and the removal of the salts, are examined in detail. Qualitative and quantitative tests are described for monitoring the removal of soluble salts and for determining the completion of the extraction process. A comparison of the quantitative expressions of salts is made.*

Introduction

Conservators are often faced with the problem of soluble salt contamination in ceramics. A study of the sources of soluble salts and their deteriorating effects on ceramics is important to an understanding of the problem and approach to treatments. The examination of the behaviour of soluble salts may be applied to many porous materials including architectural ceramics and building stones. The identification of the sources of contamination may serve to eliminate the introduction of soluble salts in some cases. The identification of salts present and a knowledge of their behavioural patterns are crucial for the comprehension of their deteriorating effects on the ceramic. In this study the discussion is restricted to ceramic objects from archaeological land sites. Marine and architectural ceramics are not considered.

The removal of soluble salts from ceramics is one of three methods suggested to prevent further deterioration. For the desalting of ceramics an identification of the salts is helpful and their quantitative determination in the extraction solution is essential. Simple means are available for their qualitative and quantitative determination by the conservator.

Sources of Soluble Salts

The contamination of archaeological ceramics by soluble salts can occur both during burial and after excavation. The sources of these salts during the burial phase is an important factor in assessing the condition of archaeological ceramics on a site as is the post-burial phase. The behaviour of the soluble salts both in the ground and in the object determines the amount of deterioration of a porous object. The most common soluble salts found contaminating archaeological ceramics are the chlorides, nitrates, phosphates, sulphates and carbonates. These salts differ in ease of solubility in water, the chlorides, nitrates and phosphates dissolving most readily. The carbonates and sulphates are often referred to as insoluble salts in the literature due to their slower dissolution in water.

In semi-arid climates and in sites near the sea chlorides may be present in substantial quantity. Sodium chloride may be carried inland from the sea in the atmosphere. Other sources of chlorides are urine and decaying flesh (Dowman 1970:29). Nitrates are formed by the oxidation of nitrogen gas, often originating from decaying organic matter (Cornwall 1958:178; Dowman 1970:29), through the conversion of ammonia by certain bacteria (USEPA 1979:4-3), or by symbiotic bacteria in plant roots by fixation of atmospheric nitrogen (Cornwall 1958:178). Phosphates are formed by the decay of organic matter (Cornwall 1958:174; Dowman 1970:29). Carbonates, present mainly as calcium carbonate, derive from calcium carbonate rocks (chalk, limestone, marble) and wood ash (Dowman 1970:23-24). Sulphates in the soil may derive from a number of sources; 1) wood ash, 2) wall plaster, 3) ammonium sulphate in modern farm fertiliser, 4) the calcium sulphates selenite and gypsum, 5) the oxidation of the sulphide minerals marcasite and pyrite, 6) the oxidation of proteins and other compounds containing sulphur (Cornwall 1958:174), and 7) sulphur dioxide in air pollution.

Fig. 1. Salt crystals hide the graffito on this ceramic.

There are three processes by which air pollutants may be introduced to the soil: 1) absorption of the gas form by moisture and vegetation, 2) deposition of the dry form as particulate matter, and 3) deposition of the wet form as precipitation (Ottar 1975:199-200). Proximity of the archaeological site to the polluting source will effect the extent to which each of these processes may occur.

Soluble salts during the post-burial phase are introduced during improper cleaning and conservation treatment. Acid is commonly used to remove incrustations of water-insoluble salts from ceramics. The object should be impregnated with water before introducing the acid in order to protect any alkaline temper in the ceramic from chemical attack and to prevent extensive migration of the acid in the pores. If the object is not wetted properly first with water the acid will be absorbed by the fabric with ensuing dissolution of the acid-soluble components in the ceramic body. The removal of the acid after cleaning will also be much more difficult. In cases where the object is impregnated with water before treatment, if the acid is not thoroughly washed away following the removal of the incrustations, it remains as a source of contaminant in the object.

Table 1
Equilibrium RH% of saturated salt solutions (From Arnold 1981:21)

Salt	*Salt compound*	*%RH*	*t°C*
chloride	KCl	84.3	25
	$NaCl$	75.3	25
	$MgCl_2.6H_2O$	33.0	25
	$CaCl_2.6H_2O$	29.0	25
nitrate	KnO_3	92.5	25
	$NaNO_3$	73.9	25
	NH_4NO_3	61.8	25
	$Ca(NO_3)_2.6H_2O$	52.9	25
	$Ca(NO_3)_2.4H_2O$	50.0	25
sulphate	K_2SO_4	97.0	25
	$NaSO_4.10H_20$	87.0	25
	$NaSO_4$	81.0	25
carbonate	$Na_2CO_3.10H_20$	92.0	18.5
	$K_2CO_3.2H_2O$	42.8	25

Hydrochloric acid may convert to chlorides (Riederer 1970:128) and nitric acid may convert to nitrates in the object (Gedye 1968:110). Chlorides and nitrates already formed during the cleaning reaction may also remain as contaminants. Pre-soaking and proper washing after treatment are important for the safe treatment and later stability of ceramics.

A source of sulphates can be the improper application of dental plaster or plaster of Paris (calcium sulphate) and the improper immersion of plastered ceramics in water during cleaning or restoration treatments. If the ceramic is not sealed or wetted properly with water before introducing the plaster, some dissolved sulphate may be drawn into the dry pores with the water solvent (resulting in a very weak plaster fill). Perhaps a greater threat of sulphate contamination comes from the partial immersion of a plastered object in water, during cleaning or restoration treatment, permitting the dissolution of the immersed plaster and the transfer by capillary action of the dissolved sulphate to the dry, unprotected areas.

Behaviour of Soluble Salts

Soluble salts may be divided into two categories by their behaviour: the anhydrous and the hydrous salts. The anhydrous salts crystallize from their solution and the hydrous salts add to their molecules water of crystallization. The chlorides, nitrates and phosphates are very soluble in water and the sulphates and carbonates are less readily soluble. Each of these is subject to a crystallization-dissolution process depending on the amount of moisture present. Each compound formed with the soluble salts has a critical point in humidity at which the transformation from the crystalline state to the dissolved state and vice versa occurs. Table 1 lists the equilibrium relative humidities of saturated solutions of various salt compounds. If the RH falls below the equilibrium RH the salts may crystallize and if the RH rises above the equilibrium RH the crystals will dissolve (Arnold 1981:20-21). The anhydrous salts are deposited as crystals upon evaporation of their water solvent under conditions of elevated temperatures and reduced moisture. In the soil these salts dissolve in water of precipitation, ground water and moisture and crystallize in the soil upon drying. In areas characterised by a hot, dry season and a cold, wet season movement of the soluble salts in the ground will occur. In cold, wet weather the anhydrous salts dissolve and are leached down into the soil. In hot, dry weather they are drawn up to the surface by capillary action as the moisture evaporates from the surface.

Fig. 2. Total-ion conductivity meter.

The migration of the hydrous salts is similar. In conditions favouring evaporation the salt solution will be drawn to the surface. The concentration of hydrous salts in solution increases as evaporation continues until over-saturation occurs, causing deposition of salts in the pores (Stambolov 1976:6). Salts which form hydrates may be characterised by different degrees of hydration. Whether or not the migrating salt reaches the surface will depend on several factors which include its distance from the surface, porosity of the object, the duration of its exposure to the alteration in RH and the degree to which this RH varies with the RH of equilibrium. In the case of the hydrous Na^2SO_4, long, needle-like crystals are precipitated on the surface. Upon continued drying if the RH falls below the RH of equilibrium for the anhydrous Na_2SO_4- 81% RH at 25°C (Table 1)- the hydrous precipitate will convert to the anhydrous, powder-like microcrystalline aggregate (Stambolov 1976:6). Hydration and dehydration of sodium sulphate may occur in the presence of water vapour according to the following reversible reaction (Arnold 1976:32):

Table 2
Qualitative and Quantitative tests

Salt	*QL*	*semi-QN*	*QN*	*Method of analysis*	*Reference*
chloride	x	x		turbidimetric	Plenderleith 1976:201
	x			spot test	Fiegl 1958:384
			x	titrimetric	Hach kit model 7-P[1]
			x	titrimetric	Hach kit model CD 50[1]
			x	colorimetric	EM Quant Test Strips[2]
			x	conductivity	total-ion meter[4]
			x	conductivity	specific-ion meter[3]
sulphate	x	x		turbidimetric	Cornwall 1958:158
	x			spot test	Fiegl 1958:408
			x	turbidimetric	Hach kit model SF-1[1]
			x	colorimetric	EM Quant Test Strips[2]
			x	conductivity	total-ion meter[4]
			x	conductivity	specific-ion meter[3]
nitrate	x			spot test	Fiegl 1958:329
			x	colorimetric	Hach kit model NI-11[1]
			x	colorimetric	Hach kit model NI-12[1]
			x	colorimetric	EM Quant Test Strips[2]
			x	conductivity	total-ion meter[4]
			x	conductivity	specific-ion meter[3]
carbonate	x			spot test	Fiegl 1958:384, 337
			x	titrimetric	Hach kit model HA-71A[1]
			x	conductivity	total-ion meter[4]
			x	conductivity	specific-ion meter[3]
phosphate	x			spot test	Fiegl 1958:385
		x			Fiegl 1958:333
			x	conductivity	total-ion meter[4]
			x	conductivity	specific-ion meter[3]
			x	colorimetry	Hach kit model PO-24[1]

$$\underset{\text{Mirabilite}}{Na_2SO_4.10H_2O + (W.V.)} \underset{hydration}{\overset{dehydration}{\rightleftarrows}} \underset{\text{Thenardite}}{Na_2SO_4 + (W.V.)}$$

(W.V. = water vapour)

Porous objects buried in the soil are subject to contamination during the dissolution and migration of salts and to subsequent physical and chemical factors of deterioration. Upon removal of a contaiminated object from damp soil to drier conditions an effloresence of salts may form on the surface. A contaminated object which is removed from a dry soil and subsequently brought into damper air can also effloresce once it dries again. The speed of crystallization of the salts is influenced by the temperature and relative humidity of the air into which the object is introduced. The greater the difference in moisture between the burial microclimate and the post-burial conditions, the greater the possibility of damage which may occur during the crystallization process. An object can even explode due to very rapid crystallization of salts (Dowman 1970:30). Once in the storage area or museum, the soluble salts contaminating the ceramic may still be subject to fluctuations in temperature and relative humidity.

Each salt compound has a threshold humidity requirement which determines whether the salt acquires a crystalline or a dissolved state. With sufficient fluctuation in RH mobility of the salt in the object can occur in the same manner as it did in the ground. With a sufficient increase in RH dissolution of the salt compounds will enable their migration in an object which has not been completely impregnated with the soluble salt. With a decrease in RH evaporation of the water will draw the salts to the surface by capillary action.

Physical and Chemical Factors of Deterioration

The damage inflicted on ceramics and other porous materials by soluble salts may be described as physical or chemical. The physical factors which cause loss of cohesion are migration, hydration, crystallization of salts and freezing. The chemical factors are hydrolysis, acidity, alkalinity, oxidation and reduction. The movement of the soluble salts during the crystallization-dissolution process and migration caused by fluctuation in RH create mechanical stress which can cause loss of the surface of the object and eventually its disintegration. Three properties of the anhydrous salts account for the breaking apart of particles and the deterioration of the ceramic body: '... they [the salts] are quite movable and thus penetrate and break many crystalline structures; they peptize i.e. suspend in water, large conglomerates of molecules; and finally augment the non-stoicheiometry of the crystals.' (Stambolov 1976:5). Due to the ease of

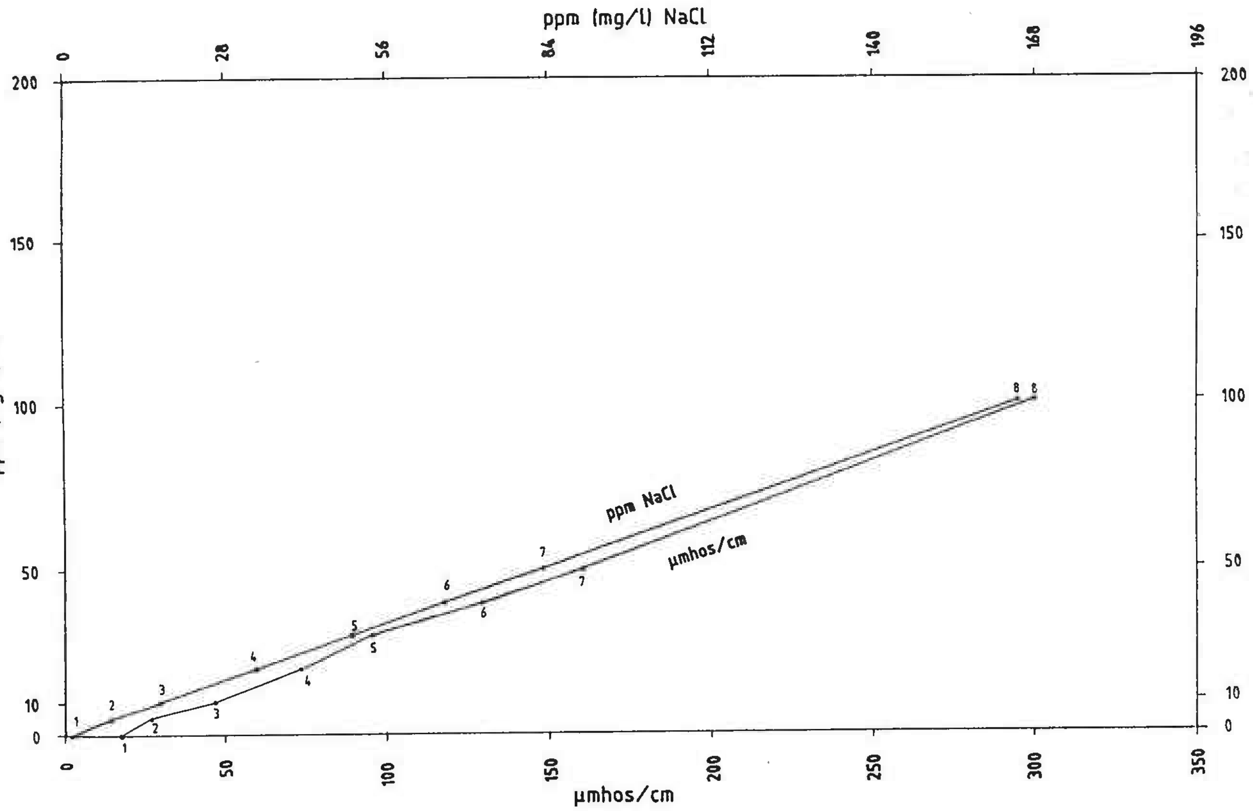

Fig. 3. Comparison of chloride ion concentration compared to NaCl concentration and conductivity

solubility in water of the anhydrous chlorides, nitrates, and phosphates, the dissolution-crystallization process occurs quickly and contributes to the extent of the damage.

More than one soluble salt compound may be present and, bearing in mind that each has its own behavioural traits, the salts influence one another. For example, nitrates are more hygroscopic than chlorides and sulphates and can lower the threshold humidity requirements for the dissolution of these salts (USEPA 1979:13-22). The hydration process may be speeded up in the presence of the anhydrous chlorides since they lower the temperature at which the transition from the hydrous to the anhydrous state takes place (Stambolov 1976:7). During the precipitation of the hydrates pressure is created in the pores which can eventually break apart the object. The onset of the disintegration creates a volume change within the pores which will determine the subsequent rate of deterioration (Stambolov 1976:7). The transformation from the anhydrous sodium sulphate to the hydrate Mirabilite creates approximately a 308% increase in volume (Stambolov 1976:6). The conversion of anhydrous calcium sulphate (Ca_2SO_4) to the hydrate gypsum ($Ca_2SO_4 \cdot 2H_2O$) is said to create a pressure of 1,100 atmospheres in the pores (Sneyers 1968:216) and the conversion of $Ca_2SO_4 \cdot {}^{1}/2\ H_2O$ (plaster of Paris) to gypsum may exceed a pressure of 2,000 atmospheres (Winkler 1970:2).

Regarding the chemical factors of deterioration, the sulphates and carbonates of calcium can cause devitrification of glazes and highly fired ceramic bodies 'by the leaching of alkalis and dissolution of colloidal silica' (Dowman 1970:27). The effects of acid materials on objects buried in the soil will depend on the concentration of the sources of the acids in the ground or in the atmosphere, buried depth of the object in the soil, porosity of the soil, buffering capacity of the soil, and ground slope (Ottar 1975:200). Sulphurous acid, formed by the hydrolysis of sulphur dioxide, and sulphuric acid, formed by the hydrolysis of sulphur trioxide, may damage objects they reach and introduce a source of sulphates in the ground. Acid causes the dissolution of limestone filler in ceramics and will deteriorate glazes which contain insufficient calcium (Dowman 1970:22).

Removal of Soluble Salts

Several conservation methods may be employed in the treatment of ceramics contaminated with soluble salts, but the removal of soluble salts is the surest method to halt and prevent any further deterioration of the ceramic body. If it is possible to identify the salt compound (s) present and maintain a stable RH for the contaminated object the deterioration process will be greatly reduced. The salts will be maintained in the object either in solution or in crystalline form although efflorescence on the surface is often unslightly and can hide detail (Fig.1). The conversion of the salt and its mobility within the object will be prevented thereby rendering the contaminant less detrimental than it would be in an uncontrolled environment with fluctuating RH. In many cases more than one soluble salt may be present and means for their identification may not be available.

Simple, qualitative tests for chlorides, nitrates, phosphates, sulphates and carbonates may be run but the determination of the salt compound may require more involved analysis (e.g. x-ray diffraction). In the cases in which the salt compounds are unidentified it is impossible to define the humidity requirement which must be respected in order to prevent conversion and movement of the soluble salts in the ceramic. If more than one salt compound is identified the determination of the humidity requirement will be difficult. In these cases

Table 3
Comparison of Units of Measurement

solution no	*Cl% wt/vol*	*Cl ppm mg/l*	*NaCl ppm mg/l*	*µmhos/cm 22 °C*
1	0.1	1	1.648	18
2	0.5	5	8.24	27
3	1	10	16.48	47
4	2	20	32.96	74
5	3	30	49.44	95
6	4	40	65.92	130
7	5	50	82.4	160
8	10	100	164.8	300

the removal of the soluble salts is the only solution to prevent further deterioration.

Some ceramics weakened by soluble salts cannot be immersed in water without ensuing damage thereby necessitating a consolidation treatment prior to salt extraction. Poulticing as an alternative method of salt extraction can be considered in these cases. Impregnation of the surface with a consolidant maybe all that is required if the salts have weakened the surface only. If more extensive damage by the salts has occured a total impregnation with a consolidant may be required. If the structural damage is so severe that consolidation of the object will not ensure its safe immersion in water, the salts may be permanently sealed in the object rather than extracted. Various problems may be associated with the consolidative impregnation of objects. The consolidant may alter the appearance of the object, may hinder the extraction of salts and may not provide sufficient protection during the salt extraction process.

The extraction process may be performed by first immersing the object in tap water, well water, rain water, etc., and ending with either distilled or deionised water. It is wise to determine the amount of soluble salt in all water used in the washing process since this can vary tremendously from location to location and can present a source of contamination itself. Methods for the extraction of soluble salts from porous bodies have been explained in detail elsewhere (Jedrezewska 1970). It is necessary to monitor the dissolved salts during the extraction process in order to determine the extent of contamination and the rate of extraction. Only by measuring the salts dissolved in the wash water is it possible to know when to stop the extraction process.

Qualitative and Quantitative Tests

Qualitative, semi-quantitative and quantitative tests for soluble salts are readily available and relatively simple to use (Table 2). Perhaps the most widely used qualitative and semi-quantitative method for determining chlorides is the silver nitrate test in which opacity of the silver chloride precipitate is the identifying means (Plenderleith 1976:201). This test is essentially qualitative although chloride concentrations may be roughly estimated by varying opacities of the silver chloride precipitate (Semczak 1977:41). A qualitative test for determining sulphates in acid solution involves the addition of barium chloride producing a white precipitate of barium sulphate (Cornwall 1958:158). It is also semi-quantitative although not as sensitive as the silver nitrate test in the 1-100 ppm range. Various proprietary kits are available for the quantitative analysis of soluble salts in which concentration is expressed in ppm (parts per million) (mg/1). The companies Hach and Merck[1]offer similar test kits. The EM Quant Test Strips by the Merck company offer a simple and quick quantitative estimation of soluble salts expressed in ppm (mg/1).[2] These are dipped in the test solution and a colour comparison made. The mineral ions potassium, magnesium, sodium, etc. may be identified as well but compounds may be difficult to ascertain if more than one is present.

The most accurate quantitative determination of salts is obtained with a specific-ion meter read in ppm such as the Orion for chlorides (Semczak 1977:40).[3] The specific-ion electrodes measure one ion to the exclusion of others which may be present.

A most convenient instrument for monitoring all soluble salts in solution is a conductivity meter for total-ionic concentration expressed in µmhos/cm or µsiemens/cm. The LaMotte Multirange conductivity meter, model DA series (Fig. 2) is one example.[4] It must be kept in mind when using these meters that more than one salt may be conducting and it is impossible to distinguish individual salts with this meter. An advantageous feature of the LaMotte meter is the temperature compensator. If cost and ease of use are determinants in the choice of test and monitoring equipment for the conservator the EM Quant Test Strips and a total-ionic conductivity meter make an excellent team at a reasonable price for the identification and quantitative estimation of soluble salts in solution.

Comparison of Units of Measurement

Depending on the type of test method used for measuring the soluble salts in solution the quantitative expression may be ppm as mg/l or moles/moles, µmhos/cm (or µsiemens/cm), or in the case of the silver nitrate test, opacity of the silverchloride precipitate. Concentration of soluble salts is sometimes given as a percentage of % as wt/vol. or wt/wt/ One example regarding porous materials states that solutions containing more than 0.1% soluble sulphates can cause damage and even efflorescence (Stambolov 1976:7). Regarding the extraction of salts from ceramics an acceptable concentration of all dissolved salts in the final wash is given as 150 µmhos/cm or less (Olive and Pearson 1975:65). Therefore it is necessary to be able to convert from one unit of measurement to another, depending upon which test method is used.

As a simple example, a comparison was made between chloride percent (% wt/vol) in solution, chloride concentration (ppm mg/l) in solution, sodium chloride concentration (ppm mg/l) in solution, and conductivity (in µmhos/cm) (Table 3). Sodium chloride solutions were prepared based on chloride concentrations as ppm (mg/l) of 1,5,10,20,30,40,50, and 100.[5]

The conductivities of these solutions were measured. The sodium chloride concentration and conductivity are compared against the chloride concentration in Figure 3. The conductivity readings increase in a linear fashion and the increases in solidum chloride concentration and conductivity closely parallel one another.

Conclusion

The importance of extracting and monitoring soluble salts cannot be overemphasized. Extensive damage can occur by the conversion and migration of soluble salt contaminants in ceramics and other porous objects in a climate with uncontrolled relative humidity. A quantitative analysis of the salt contaminants will dictate the removal of salts if necessary. Monitoring the soluble salts during the extraction process is essential in order to define a safe completion of the washing process. Overwashing of ceramics can leach out soluble compounds which will weaken the fabric and is one more reason to monitor the extraction process.

Only one published reference has been cited regarding an acceptable concentration of soluble salts in solution as an indication of the completion of the extraction process (Olive and Pearson 1975:65). It is not mentioned if the figure of 150 µmhos/cm was derived arbitrarily or if it is based on the testing and examination of objects whose salts were extracted according to this criterion.

Preliminary tests indicate that a safe level of chloride concentration in the wash water is 75-1000 µmhos/cm. Ceramic lamps which were contaminated predominantly by chlorides have been desalted within the past six years to readings of 75-100 µmhos/cm in the wash water.[6] These lamps have been stored in uncontrolled environmental conditions since their desalting and have not displayed salt effloresence or further deterioration.

More extensive study is required to determine safe levels for the various soluble salts in the extraction solution. An acceptable concentration derived by hydration-dehydration testing of objects, which have been desalted according to known concentrations of salts in the extraction solution, will be based on the absence of deterioration caused by salt conversion and migration.

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Notes

1. Hach Chemical Company, World Headquarters, P.O.Box 389, Loveland, Colorado 80537, USA. Hach Europe S.A./N.V., B.P. 51, 5000 Namur 1, Belgium.
2. EMQuant Test Strips, EM Science, 111 Woodcrest Road, P.O.Box 5018, Cherry Hill, New Jersey 08034, USA. E. Merck, Frankfurter Strasse 250, D6100, Darmstadt 1, W. Germany.
3. Orion specific-ion meter and electrodes available from Sargent-Welch Scientific Company, 7300 North Linder Avenue, P.O.Box 1026, Skokie, Illinois 60077, USA.
4. LaMotte Multirange conductivity meter; LaMotte Chemical Products Company, P.O.Box 329, Chestertown, Maryland 21620, USA.
5. Prepared and measured at the Central Institute of Restoration, Rome.
6. Agora Excavation and Museum, American School of Classical Studies, Athens, Greece.

References

Arnold, A. (1976) 'Behaviour of Some Soluble Salts in Stone Deterioration', *Second International Symposium of the Deterioration of Building Stones,* 27-36.

Arnold, A. (1981) 'Nature and Reactions of Saline Minerals in Walls', *The Conservation of Stone II,* 13-23, Proceedings of the International Symposium, Bologna,

Cornwall, I.W. (1958) *Soils for the Archaeologist,* Phoenix House Ltd., London.

Dowman, E.A. (1970) *Conservation in Field Archaeology,* Methuen & Co. Ltd., London.

Fiegel, F. (1958) *Spot Tests in Inorganic Analysis,* Elsevier Publishing Co., New York.

Gedye, I. (1968) 'Pottery and Glass', *The Conservation of Cultural Property,* 109-113, UNESCO, Paris.

Jedrzejewska, H. (1970) 'Removal of Soluble Salts from Stone', *Conservation of Stone* 1, 19-33, IIC, New York.

Olive, J., Pearson, C. (1975) 'The Conservation of Ceramics from Marine Archaeological Sources', *Conservation in Archaeology and the Applied Arts,* 199-204, IIC, Stockholm.

Ottar, B., Haagenrud, S.E. (1975) 'Air Pollution and Possible Effects on Archaeological Objects Buried in the Ground', *Conservation in Archaeology and the Applied Arts,* 199-204, IIC, Stockholm.

Plenderleith, H.J., Werner, A.E.A. (1976) *The Conservation of Antiquities and Works of Art,* Oxford University Press, London.

Riederer, J. (1970) 'Stone Preservation in Germany', *Conservation of Stone* 1, 125-134, IIC, New York.

Semczak, C.M. (1977) 'A Comparison of Chloride Tests'. *Studies in Conservation* 22, 40-41.

Sneyers, R.V., De Henau, P.J. (1968) 'The Conservation of Stone', *The Conservation of Cultural Property,* 209-235, UNESCO, Paris.

Stambolov, T. (1976) 'The Corrosive Action of Salts', *Lithoclastia* 1, 3-8.

United States Environmental Protection Agency (1979) *Air Quality Criteria for Oxides of Nitrogen,* USEPA: Research Triangle Park.

Winkler, E.M. (1970) 'Decay of Stone', *Conservation of Stone* 1, 1-14, IIC, New York.

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