Identification of Lime Plaster in Prehistory Using Petrographic Methods: A Review and Reconsideration of the Data on the Basis of Experimental and Case Studies

Panagiotis Karkanas*

Ephoreia of Palaeoanthropology-Speleology of Southern Greece, Arditto 34b, 11636 Athens

Most prehistoric plasters and mortars consist of very small amounts of burnt lime mixed with anthropogenic debris, soil, and sediment. To solve the problem of identification of such small amounts of lime in impure lime plasters, a series of experimental plasters were prepared and studied with petrographic methods. Samples of living floors from five prehistoric sites in Greece were also reanalyzed under the light of the experimental findings and compared with natural calcareous sediment. The most promising features for identifying lime are transitional textures of partially carbonized slaked lime that can be observed in the lime lumps and the binding matrix. They are usually in the form of ill-crystallized portlandite and calcite mixtures or cryptocrystalline calcite. Well-reacted calcitic groundmass, shrinkage fractures, and occasionally colloidal forms are also additional indications. The present experimental study shows that lime could be easily produced by heating porous, soft calcareous materials. This would probably account for the very frequent use (in small quantities) of lime in Greek prehistory. Plaster and mortars were made by mixing damp anthropogenic dirt as aggregate and fragments of quicklime, a technique known as “hot mixing.” © 2007 Wiley Periodicals, Inc.

INTRODUCTION

Although considerable interest has been expressed in the production of burnt lime in prehistory, relatively little has been written on its identification problems, which are particularly evident in old, weathered, and impure samples (cf. Affonso, 1996). For example, Kingery et al. (1988) identified lime in a plastered floor from Çatalhöyük, using scanning electron microscopy (SEM); but Matthews et al. (1996), using standard optical petrographic methods, suggested that the plastered floors of Çatalhöyük were prepared with unburnt soft calcareous material. Affonso (1996) has pointed out that matrix burnt lime can be very similar to unburnt fine calcareous sediment. Indeed, the recarbonation process of slaked lime (lime putty) produces calcium carbonate (calcite) that minerallogically and chemically is indistinguishable
from raw sedimentary calcite (Gourdin and Kingery, 1975; Goren and Goldberg, 1991; Moorey, 1994: 330). Isotopic values of oxygen can be sufficient for differentiating burnt lime (Zouridakis et al., 1987), but this method is not a routine procedure. Moreover, this and similar methods are bulk methods that cannot answer the question whether burnt lime was added accidentally as inclusions in the plaster or represents true cementing binder, particularly in samples where lime was used in small quantities (see, for example, Affonso and Freiberg, 2001).

Scanning electron microscopy was one of the first methods employed in the study of prehistoric lime (Gourdin and Kingery, 1977; Kingery et al., 1988), but it is suitable only for point observations, because larger scale features and the geometric relationships—that is, fabric—of the matrix and the aggregates are not readily discerned (c.f. Barnett, 1991, contra Kingery, 1991). Petrographic microscopy seems to be the best analytic tool for prehistoric lime plaster, because both the mineralogy and the fabric are studied on undisturbed samples (Goren and Goldberg, 1991, Affonso and Freiberg, 2001). Indeed, petrographic analysis is still one of the major tools in the study of historical mortars where information on the binder complexity, lime inclusions, nature of aggregates, and pore structure are increasingly important in the conservation of buildings (Güleç and Tulun, 1997; Hughes et al., 2001; Degryse et al., 2002; Leslie and Hughes, 2002; Macphail, 2003). However, in the case of historic lime mortar and plaster there is no question on the nature of lime, because the amount of used burnt lime was enough to produce a distinct fabric and to impart an obvious high mechanical strength to the material. In contrast, prehistoric lime plaster seems to contain very low and variable amounts of burnt lime (if not at all) and is mixed with a diverse range of materials including clay, dung, unburnt limestone and chalk, soil, everyday anthropogenic debris, etc. (e.g., Goren and Goldberg, 1991, Affonso and Freiberg, 2001). Hence, the evidence of lime presence in the end product is very weak. The identification of lime use in prehistoric sites is of major interest, not only for understanding the technological modifications associated with the emergence of complex societies, but also for the ensuing economic and social implications like craft specialization, labor intensification, and resource management (Garfinkel, 1987; Kingery et al., 1988; Goren and Goldberg, 1991).

A series of ongoing studies in prehistoric sites in Greece revealed that burnt lime was indeed used in several cases (Karkanas, 2002; Karkanas and Efstratiou, 2003). In addition, several “mud” plastered floors were closely associated with unequivocal lime plastered floors implying, probably, the existence of a continuum where the distinguishing line between presence and absence of lime is diffuse. It was also realized from these ambiguous cases that the use of small quantities of lime could be more frequent than previously thought. Therefore, a research project was undertaken that aimed at resolving some of the problems of identification of lime in prehistoric contexts, mainly through observations of petrographic thin sections. I prepared experimental lime plastered floors using different mixtures and quality of lime with archaeological dirt, and I reanalyzed the prehistoric plastered floors using the experimental data. Using this data, it was possible to refine some of the identification criteria for the presence of lime.
IDENTIFICATION OF LIME PLASTER IN PREHISTORY

METHODOLOGY

Experimental Plasters

An experimental study was carried out to observe the different structures and the mineralogical changes associated with the preparation of lime plasters. For the production of quicklime (CaO), a hard massive dense microsparitic limestone and a soft porous pisolite calcareous tufa (fluvial travertine) were used (Table I). The calcareous tufa was sampled from the vicinity of one of the studied sites (Makri, Thrace, Table II), and the hard limestone from the Parnassos carbonate nape unit in central Greece, an area where lime was produced traditionally until a few decades ago. A series of lime samples were prepared by burning fragments <5 cm of the limestone samples from 1 to 7 hours at 800 and 1000 °C (Table I). Plaster test specimens were manufactured in a variety of ways. Pure lime plasters were prepared inside aluminum moulds by adding small amounts of water into the quicklime fragments until no reaction was observed. For the impure lime plasters, different proportions of sediment (obtained from the excavation at Makri, one of the studied sites) were used (Table II). The sediment was a mixture of anthropogenic debris, clay, and calcareous clastic sediment. The calcareous clasts comprised mostly tufa fragments that were also used for the production of one of the lime types. The preparation of most of the lime plasters followed the hot mixing procedure (dry slaking); unslaked quicklime was mixed directly with aggregate and enough water to work the material in situ, but lime putty (slaked lime) was not prepared in advance (cf. Hughes et al., 2001; Leslie and Hughes, 2002). This method was preferentially used because there were strong indications that it was the one used in the studied sites. However, two specimens were prepared by using industrial, pure, homogeneous lime putty. Plasters without lime were also prepared by mixing sediment and water. In one of the experiments overburnt (dead) lime was produced (see Table I). The dead lime was immersed in water for a month and then it was let to dry inside the mould. All plasters were left to carbonate one month before petrographic thin sections were constructed.

Table I. Experimental plasters using mixtures of aggregate and different types of lime.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Temp. (°C)</th>
<th>Time (hours)</th>
<th>Lime raw material</th>
<th>Aggregate</th>
<th>Lime reactivity</th>
</tr>
</thead>
<tbody>
<tr>
<td>exp1</td>
<td>1000</td>
<td>7</td>
<td>microsparitic limestone</td>
<td>–</td>
<td>overburnt lime</td>
</tr>
<tr>
<td>exp2</td>
<td>1000</td>
<td>3</td>
<td>microsparitic limestone</td>
<td>–</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp3</td>
<td>1000</td>
<td>3</td>
<td>microsparitic limestone</td>
<td>25%</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp4</td>
<td>1000</td>
<td>3</td>
<td>microsparitic limestone</td>
<td>35%</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp5</td>
<td>800</td>
<td>2</td>
<td>microsparitic limestone</td>
<td>–</td>
<td>slightly reacted</td>
</tr>
<tr>
<td>exp6</td>
<td>800</td>
<td>2</td>
<td>microsparitic limestone</td>
<td>50%</td>
<td>slightly reacted</td>
</tr>
<tr>
<td>exp7</td>
<td>800</td>
<td>2</td>
<td>tufa</td>
<td>–</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp8</td>
<td>800</td>
<td>2</td>
<td>tufa</td>
<td>25%</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp9</td>
<td>800</td>
<td>2</td>
<td>tufa</td>
<td>50%</td>
<td>readily reacted</td>
</tr>
<tr>
<td>exp10</td>
<td>800</td>
<td>1</td>
<td>tufa</td>
<td>–</td>
<td>readily reacted</td>
</tr>
<tr>
<td>MKDAL</td>
<td>n.a.</td>
<td>n.a.</td>
<td>industrial lime putty</td>
<td>50%</td>
<td>homogeneous slaked</td>
</tr>
<tr>
<td>MKD</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>100%</td>
<td>–</td>
</tr>
<tr>
<td>MKDA</td>
<td>–</td>
<td>–</td>
<td>–</td>
<td>100%</td>
<td>–</td>
</tr>
</tbody>
</table>

n.a. = not available.

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Table II. Studied lime plasters from archaeological sites.

<table>
<thead>
<tr>
<th>Sites</th>
<th>Field description</th>
<th>Microscopic description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Makri, Thrace, Northern Greece</td>
<td>Sequences of whitish hard floors and brownish gray compact layers (thickness of individual layers 1–5 cm).</td>
<td>Pure lime plastered floors with large amounts of lime lumps and half-burnt tufa remnants, impure lime floors consisting of a mixture of burnt lime, varying amounts of anthropogenic debris (ash, charcoal, bone, dung, plant remains), and siliciclastic sediment.</td>
</tr>
<tr>
<td>Drakaina Cave, Kefallonia, Ionian Islands</td>
<td>Whitish hard plaster floors.</td>
<td>Almost pure lime plastered floors with large amounts of lime lumps, half-burnt fossiliferous marl remnants, and occasional clay admixtures.</td>
</tr>
<tr>
<td>Palamari, Skyros Island, central Aegean</td>
<td>Thinly layered sequences of reddish and greenish compact sandy loam (thickness of individual layers 1–10 cm) from both sides of a fortification wall.</td>
<td>Laid floors of each time unique mixture of angular mica schist detrital minerals, rounded quartz sand of aeolian origin (aeolianite parent rock), clay, and small amounts of burnt lime.</td>
</tr>
<tr>
<td>Mitrou islet, Atalanti, central Greece</td>
<td>Living surfaces of gray brown, homogeneous, and moderate loose sandy loam with occasional whitish calcareous lenses.</td>
<td>Mostly highly bioturbated anthropogenic rich sediment with some preserved lenses of impure lime plasters. The latter consist of a mixture of serpentinite sand, anthropogenic debris, and small amounts of burnt lime with half-burnt calcareous marl fragments.</td>
</tr>
<tr>
<td>Geraki, Peloponnese, southern Greece</td>
<td>Living surfaces of homogenous compact reddish brown sandy loam.</td>
<td>Mixture of red decalcified clay, burnt dung, and burnt lime with fragments of half-burnt calcareous schist.</td>
</tr>
</tbody>
</table>
Archaeological Samples

The studied samples come from five prehistoric sites in Greece (Makri, Palamari, Mitrou, Drakaina, and Geraki) (Table II) ranging from Late Neolithic (6th millennium B.C.E) to Late Bronze Age (11th century B.C.E) (Parlama, 1994; Chatziotou et al., 1995; Efstratiou et al., 1998; Crouwel et al., 2005; Van de Moortel and Zahou, 2006). Micromorphological studies have been undertaken in all these sites, so the studied material was already available. In some cases, the samples were already recognized in the field as possible lime plastered floors on the basis of their unusually mechanical strength, but in others, they were not even considered as plastered floors. However, all samples came from house interiors where several archaeological indications pointed to possible presence of living surfaces. Under the microscope it was possible to identify them as floors based on their microstructure and form. The presence of a dense homogeneous groundmass with random distribution of coarse components and the frequent observation of vughs and vesicles are the most indicative microscopic features of floors (for a review, see Ge et al., 1993, and Matthews, 1995). However, in most cases an additional decisive criterion was the presence of a clear-cut smooth upper surface, and sometimes a finishing coating (Figure 1).

**Figure 1.** Superimposed thin plastered floors with sharp contacts and finishing coats (Markri, sample MK408a, height of photo [h.p.]: 1 cm).
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For comparative reasons, three more samples from different late Byzantine mortars from my reference collection were included in the study.

Other Reference Samples

In all studied sites, the calcareous rocks that were identified as possible sources of lime production were also sampled and studied microscopically. In addition, comparative studies were carried out on soft calcareous lacustrine sediment (from the Marathon plain and Karla Lake), travertines (from Makri and several caves), and calcrete soils (from the Nemea Valley) that were available in my reference collection.

Both large (5 × 7.5 cm) and regular format (4.5 × 2.5 cm) thin sections were prepared by using standard micromorphological techniques that involve impregnation of the samples with polyester resin diluted with styrene (Murphy, 1986).

RESULTS

Production of Quicklime

All but two experiments produced readily reacted quicklime (Table I). Except for the overburnt lime (hard limestone, 7 hours at 1000°C) that slaked poorly, an insufficiently burnt lime was produced by heating the hard limestone at 800°C for 2 hours. In this experiment about 70% of the limestone was not transformed to quicklime (visual estimation on thin section). In contrast, the soft calcareous tufa produced almost completely burnt lime (about 80%) at 800°C in only 1 hour.

Experimental Plasters

The experimental pure lime plasters contained three different types of lime: binding matrix, lime lumps, and half-burnt limestone fragments.

The cores of the half-burnt limestone fragments in the case of the hard limestone show only slight changes. The tendency of limestone to become brownish gray when heated (see Courty et al., 1989: 109) was confirmed (Figure 2). In addition, the boundaries of the crystals and particularly the cleavage planes became more discernible and attenuated as clear black lines, most likely because they are the areas where the carbonization front advances (Figure 2). However, all these changes cannot be considered unequivocal signs of burning, because they overlap with weathering alterations observed in parent limestone rocks. Nonetheless, a unique, but infrequent feature that is observed in heated limestone fragments is the formation of dark gray to black isotropic patches having a reticulate or glomerated radial pattern due, most likely, to incipient internal fissuring (Table III; Figure 3).

Nevertheless, where heating was more intense, the half-burnt limestone fragments show edges and rims of reacted lime (Figure 2). These reaction rims are particularly diagnostic, because they are not encountered in natural sediment (Table III). They consist of different types of reacted lime observed also in the matrix and in the lime lumps described below.
Lime lumps are the dominant features of all lime plasters. They have several origins like overburnt quicklime, unreacted or half-reacted quicklime, and crudely slaked or mixed lime (Leslie and Hughes, 2002). Even in the case of the industrial putty lime, insufficient mixing produced some lime lumps (Figure 4a). Hot mixing preferentially produces lime lumps because of the low amount of water used (poor slake) and the crude mixing with the aggregate.

In the experimental specimen, lime consisting of poorly reacted quicklime tends to be colorless or light beige and has very low relief; in cross-polarized light it is mostly isotropic (Table III; Figure 5). Areas with incipient reaction are dominated by dispersed, very fine (almost unresolved), ill-crystallized, colorless portlandite (Ca(OH)$_2$) with very low relief. This type of portlandite has white or low-order gray birefringence (Figure 6). However, large platelets of portlandite with typical first-order blue to second-order red and occasionally perfect basal cleavage were also observed (see also review of portlandite microscopic appearance in St. John et al., 1998: 99–100 and 381–382). The large crystals of portlandite are more rare and

Figure 2. Half-burnt limestone fragment with a brownish (medium tones of gray in photo) very low relief rim of slaked lime showing some banding. The core has become locally dark gray, with cleavage plains clearly discernible as black lines. Shrinkage fractures crosscut the whole fragment (sample exp5, plane polarized light [PPL], h.p.: 2 mm). Insert shows part of a half-burnt limestone fragment from Makri that has also become darkened. (MK408, PPL, h.p.: 1.2 mm).
<table>
<thead>
<tr>
<th>Observations</th>
<th>Microscopic features of lime</th>
<th>Experimental plaster</th>
<th>Archaeological samples</th>
<th>Natural sediments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reticulate and glomerate structures in lumps</td>
<td>black to dark gray, moderate</td>
<td>half-burnt limestone</td>
<td>few</td>
<td>n.o.</td>
</tr>
<tr>
<td>Amorphous lumps and matrix</td>
<td>colorless, very low</td>
<td>amorphous portlandite</td>
<td>common, n.o.</td>
<td>n.o.</td>
</tr>
<tr>
<td>Partly amorphous lumps and matrix</td>
<td>colorless, very low</td>
<td>ill-crystallized portlandite</td>
<td>common, very few, very few</td>
<td>n.o.</td>
</tr>
<tr>
<td>Crystal platelets with occasionally basal cleavage</td>
<td>colorless, very low</td>
<td>portlandite</td>
<td>few, n.o.</td>
<td>n.o.</td>
</tr>
<tr>
<td>Semi-amorphous lumps, or rims around lumps and matrix</td>
<td>beige to light brown, moderate</td>
<td>mixture of ill-crystallized portlandite and calcite</td>
<td>common, frequent, few</td>
<td>n.o.</td>
</tr>
<tr>
<td>Semi-amorphous to crypto-crystalline matrix and lumps</td>
<td>dark brown to golden brown, moderate</td>
<td>mixture of ill-crystallized portlandite and calcite</td>
<td>frequent, few, few</td>
<td>n.o.</td>
</tr>
<tr>
<td>Cryptocrystalline lumps and matrix</td>
<td>dark gray, moderate</td>
<td>crypto-crystalline micritic calcite</td>
<td>frequent, frequent, to common</td>
<td>common in biogenic limestones and travertines</td>
</tr>
<tr>
<td>Crystalline lumps and diffuse patches in matrix</td>
<td>light gray, moderate</td>
<td>micritic calcite</td>
<td>few, common</td>
<td>common</td>
</tr>
<tr>
<td>Crystalline matrix and lumps</td>
<td>colorless</td>
<td>moderate</td>
<td>high-order pale colors</td>
<td>micritic calcite</td>
</tr>
<tr>
<td>----------------------------</td>
<td>----------</td>
<td>----------</td>
<td>------------------------</td>
<td>-----------------</td>
</tr>
<tr>
<td>Fractures with hierarchical pattern</td>
<td></td>
<td></td>
<td></td>
<td>shrinkage fractures</td>
</tr>
<tr>
<td>V-shaped and lenticular voids in lumps</td>
<td></td>
<td></td>
<td></td>
<td>shrinkage voids</td>
</tr>
<tr>
<td>Curvilinear or concentric voids in lumps</td>
<td></td>
<td></td>
<td></td>
<td>shrinkage voids</td>
</tr>
<tr>
<td>Colloidal forms in lumps</td>
<td></td>
<td></td>
<td></td>
<td>crystallization of colloidal slaked lime</td>
</tr>
</tbody>
</table>

n.o. = not observed

1 All archaeological samples invariably contain amounts of clastics and anthropogenic debris.

2 Note that the relief of crystalline calcite is moderate, but on rotation of the stage changes markedly. However, this can rarely be observed in micritic calcite.

3 With variable amounts of elastic sediment and anthropogenic debris.
tend to arrange along the edges of the lime lumps or inside fractures and voids (Figure 5).

A white, carbonated zone extending about half a centimeter from the surface of the experimental plasters is easily defined macroscopically. At the advancing front, it crosscuts the lime lumps (Figure 7). Inside this zone the lime has a strong brown color and low to moderate relief; it consists of very fine unresolved crystallites with golden brown or first-order dull blue and reddish birefringence colors (Table III; Figures 5 and 7). Occasionally the birefringence becomes of high order, revealing better crystallized, but still very fine, calcite. In other cases the lime lumps are totally featureless dark gray to dark brownish-gray that also mask the birefringence colors (Figure 5). The latter feature is typical of cryptocrystalline micrite observed in several types of natural carbonate sediments (Table III). Characteristic of this carbonation front are lime lumps with some concentric zoning presumably related to fluctuations in hydration or carbonation rate (Figure 7). Small subangular platy lumps having remnants of zoning are detached from the larger ones.

Figure 3. Dark isotropic features in half-burnt limestone: (a) Reticulate pattern in experimental lime plaster. Note the rounded vesicle in the lower part of the photo (exp11, PPL, h.p.: 2 mm); (b) globular, rosette-like structure (Makri, MK201, PPL, hp: 1.3 mm); and (c) globular pattern (Palamari, PL5a1, PPL, hp: 0.7 mm).
The addition of low amounts of sediment aggregate (<0.3:1) seems not to affect the overall appearance of the lime plaster. In contrast, the addition of larger amounts of sediment (1:1) has some interesting consequences on the carbonation process of lime. Very few areas of the lime matrix or lime lumps are not carbonized. These areas have still a semi-isotropic groundmass with a hazy, pixilated, white-gray appearance in cross-polarized light (Table III; Figures 4a and 6). There are also dark gray to almost black, featureless cryptocrystalline areas in the matrix where the sediment aggregates are embedded, and light gray, sand-sized areas with dense high birefringence micrite. The latter are presumably small lime lumps totally carbonized and well incorporated inside the crystallized lime matrix (Figure 4a).

All experimental lime plasters are characterized by the presence of a complex shrinkage fracture system inside both the lime lumps and the matrix (Table III; Figures 2, 5–7). Typical shrinkage fractures have a very irregular, dense zig-zag appearance. They also exhibit a somewhat hierarchical pattern, generating a reticulate network.

Figure 4. (a) Experimental lime plaster (MKDAL, PPL, h.p.: 1 mm). Gray, sand-sized, carbonized lime lumps of micritic calcite embedded in moderate carbonized matrix; also areas of low relief not readily discerned (light gray areas) and lumps with dark featureless appearance (right side of photo); (b) lime plaster floor from Makri (MK405b, PPL, h.p.: 1 mm). The left side of the photo is dominated by dark featureless groundmass that gradually passes to a light gray groundmass with embedded, gray, sand-sized lime lumps (in XPL the left side is isotropic and the rest is highly birefringent).
of pores; their walls are accordant surfaces. Sometimes they have a propagation center (radial cracks) and shattering edges. In addition, lime lumps have lenticular, arcuate, and v-shaped form of voids. It also seems that, as carbonization proceeds, fractures tend to be less angular, attaining curvilinear and sometimes concentric patterns (Figure 7). These are attributed to the crystallization of the hydrated colloidal slaked lime, the density of which depends on the water content (Stefanidou and Papayianni, 2005). In the case of the impure lime floors, rounded vesicles and irregular voids with smooth curved walls are also observed (Figure 3a). Shrinkage fractures are one of the unique characteristics of lime lumps and lime matrix and have been reported in the literature (Leslie and Hughes, 2002, Hughes et al., 2001, Elsen et al., 2001, Shahack-Gross et al., 2005). Although sometimes they are present in large amounts, the scale of the fractures does not seem to affect the structural integrity of the plaster (see Leslie and Hughes, 2002).

Plaster floors without lime have a similar compact and dense appearance under low magnifications; they are very difficult to differentiate from the impure lime plaster specimens with large amounts of dirt (1:1). However, at higher magnifications,
the matrix, although sometimes dense, does not show signs of reaction. That is, the individual crystallites of calcite do not form a continuous matrix with interconnected boundaries but are usually isolated or embedded in a clayey matrix. The plasters are not characterized by shrinkage fractures, but only by isolated fractures and vugly to vesicular porosity. Although calcareous aggregate is a major component of these plasters, they do not show most of the described characteristics of lime lumps. In most of the cases, the boundaries with the matrix are clear and there are no signs of reaction with it.

Archaeological Samples

The archaeological samples that have an unusually high mechanical strength (and were tentatively identified in the field as lime plasters) are the ones with the clearer signs of burnt lime. They are all characterized by a high amount of lime lumps that are well reacted with the surrounding matrix. There is always a diffuse halo of micrite around the lumps that cannot be differentiated from the matrix. Amorphous halos with a hazy, pixilated, white-gray appearance under cross-polarized light were observed (Table III; Figures 8a). Some lumps have nonburnt or half-burnt cores,
recognized by the preservation of structures found in the parent rock materials (biofabrics, fossils, equigranular sparite, etc.) (e.g., Figure 8b). Most of the features observed in experimental impure lime plasters were also observed in the archaeological samples (Table III; Figures 8-12). However, in most cases, the lumps consist of very fine, light gray crystallites of calcite with high birefringence colors, presumably due to the complete carbonization (Figure 4b, 8c, 9, and 10).

Lime binding material has the typical appearance of continuous reacted fine calcitic matrix (Figure 11). Dark gray to almost black featureless areas with embedded non-limy aggregates are also observed (Figure 4b). Rarely, there are areas in the matrix with low relief and hazy, pixilated, low-order birefringence color (Figure 12). They are probably a mixture of ill-crystallized portlandite and calcite.

Shrinkage fractures, rounded vesicles, and irregular vughs with smooth and curved walls are also found (Figures 8b, 9, and 10). Shrinkage fractures are more often observed in poorly carbonized lime lumps. In addition, a pronounced feature that was observed in the case of Makri lime plastered floors was the presence of pelletal

Figure 7. Carbonization front in experimental lime plaster (exp4, XPL, h.p.: 2 mm). Note the banded appearance of the moderate brown (dark gray in photo) birefringent carbonized part of the lime lump (a), in contrast with the dark, isotropic, poorly carbonized other half (b). Note also the lower part of the same lime lump that has a gray-speckled, low birefringent color (c). In PPL, this lower part has a light beige color and low relief, whereas the upper parts are dark brown and moderate relief. Shrinkage fractures are evident with locally concentric patterns (d).
lime matrix structures (Figure 8d). The pellets consist of fine gray, high birefringent calcite; have variable sizes and shapes; and laterally pass to dense lime matrix. Pelletted textures have been observed in lime lumps (Hughes and Leslie, 2001), and pelletal concentrations of lime between aggregate grains, described as meniscate cement, are reported by Leslie and Hughes (2002).

In very few cases, a macroscopic whitish carbonation front was observed advancing from the plaster surface (as at Drakaina cave). Under the microscope, platy lime lumps with some internal banding, reminiscent of the ones observed in the experimental specimen, were recognized (Figure 8c).
Even the purest lime prehistoric plasters contain small amounts of anthropogenic debris in the form of clastic sediment mixed with burnt remains (charred material and ash). However, in these cases, the lime lumps play the role of aggregate and give a mortar appearance to the material.

DISCUSSION

Prehistoric plasters and mortars that contain high amounts of well-reacted lime are easily identified by their dense, highly birefringent crystallitic b-fabric of the reacted lime binding material (e.g. Courty et al., 1989:121 and Plate IVd). This type of matrix was frequently confirmed in the archaeological lime plaster samples studied in this work (Figure 11). The archaeological samples differ from the experimental specimen in that the latter are dominated by transitional textures where carbonation of the slaked lime is not completed. Nevertheless, transitional textures were also observed, though less frequently, in the archaeological samples; they are the most diagnostic of the presence of lime in ambiguous cases (Figures 4b and 8-12). Transitional textures, particularly the hazy, gray-white, semi-isotropic appearance of the lime matrix, were also observed in all Byzantine mortar samples.
Samples with progressively higher amounts of aggregate in the form of anthropogenic dirt and soil tend to resemble mud plasters with no lime. Surprisingly, however, in most of the cases where a plaster floor was identified in the studied sites, careful observation revealed that they also contained some lime. The most persistent and easily identified criteria are lime lumps with shrinkage fractures and reaction rims with the matrix (Figures 8a-c and 9); lime matrix with dark gray, featureless, isotropic to cryptocrystalline appearance (Figure 4b); and light brown matrix with hazy, pixilated, white-gray birefringence colors (Figure 12). One of the most difficult criteria to describe and identify is the presence of calcitic-reacted matrix in the presence of other clastic material. However, there are always patchy areas with denser crystallitic b-fabric that also have embedded gray, sand-sized lumps of micritic calcite (see Figures 4b and 9).

Several of the features observed in the experimental lime specimen were also observed in natural calcareous sediment. For example, fragments of micritic limestone (and especially travertines, calcrites, and lacustrine marls) can have a dark gray or brownish-gray, featureless, cryptocrystalline appearance in plane polarized light and moderate-to-low-order brownish colors in cross-polarized light. However, complex shrinkage fractures were never observed. Care should be taken, though, with...
some pore structures associated with calcretes, travertines, and general microbial carbonates. In these cases voids tend to be isolated fractures or equant in shape and elongated parallel fenestrae (c.f. Tucker, 2003: 120; Pentecost, 2005: 30–31). It was also observed that the carbonization process of lime lumps tend to seal the shrinkage fractures and leave some residual, fenestrae-like voids that could be misidentified as natural formed pores. However, in most of the cases, lime voids have smooth curved walls and embayed forms (Figure 10). Lime lumps in very impure lime plasters have not always reacted with the matrix, because the latter does not contain sufficient lime to become the binding agent. In these cases, even well-crystallized lime lumps have somewhat curvilinear and globular edges; they are associated with meniscate voids (Figure 10) that are attributed to crystallization of the hydrated colloidal slaked lime.

Due to the high amounts of clay, clastic calcite, ash crystals, and other fine-grained material, lime matrix in highly impure lime plasters is very difficult to differentiate from impure clastic, micritic, or biogenic lacustrine and from similar calcareous sediment. Biogenic calcareous sediments do contain reacted calcite structures (actually chemically or biogenically precipitated), but if present in a plastered floor, these structures should be preserved only in the aggregate and not in the binding matrix.
Therefore, identifying the nature and texture of the binding matrix is of major importance in plastered floors.

To recognize small amounts of lime, it is also crucial to identify first the sample as part of a plastered floor or similar construction material. Indeed, secondary calcite cementation in open areas tends to mimic the reacted calcareous matrix of lime plastered floors. There is also the possibility of secondary calcite cementation of exposed sequences of floors, but, normally, it crosscuts floor contacts and is also associated with root passages or sometimes root alveolar structures.

The very large amounts of poorly slaked lime lumps found in the archaeological samples strongly suggest that the plasters were made by mixing damp aggregate with crushed quicklime. It has been postulated that these types of mortars, known as “hot lime” mixes, appear to have better setting and bond properties than mortars made by mixing lime in the form of putty (Moropoulou et al., 1996; Leslie and Hughes, 2002). In the present experimental specimen, there were indications that the addition of considerable amounts of mixed soil and anthropogenic dirt improves the carbonation process. Moreover, this technique does not require well-burnt lime, because even half-burnt limestone can play the role of lime lumps and give a mortar texture to the material.

Figure 12. Detail of an impure lime plaster floor where the hazy, pixilated, gray-white areas consist of a low-birefringent, low relief, and light beige (in PPL) mixture of ill-crystallized portlandite and calcite (Makri, MK26a XPL, h.p.: 2 mm).
Goren and Goldberg (1991), based on the petrography of lime plastered floors in the Neolithic of the Near East, suggested that lime burning was a casual, limited activity not requiring intensive labor. The results of the present study strongly confirm this conclusion. In all studied cases, insufficient burning is evident, and lime was mixed with unpicked aggregate collected most likely from the streets or the immediate surroundings of the settlements. The raw material that was used in the studied cases was soft marl (Drakaina Cave and Mitrou), calcareous tufa (Makri), friable calcareous schist, and calcrete (Geraki) and calcareous aeolianite (Palamari). In all cases, the material could be easily crushed in small fragments, thus reducing the temperature of quicklime production. As shown in the experimentally produced quicklime and reported in the literature (Moropoulou et al., 2001), the soft porous nature of most of these materials enhances their reactivity and lowers the temperature of burning. For tufa travertine, only one hour at 800 °C was sufficient to transform most of it to readily reacted quicklime. It is expected, however, that the firing condition of the same limestone will be different in open kilns like the ones probably used in prehistory. In any case, it seems that small amounts of moderately well-burnt lime can be produced easily in a few hours, using reasonable amounts of wood as fuel (see also Affonso and Freiberg, 2001). If this were the case, it would probably account for the scale of use of lime in the Greek prehistory. Lime was used very frequently, but in small quantities.

CONCLUSIONS

Transitional textures of partially carbonized slaked lime represent the most secure evidence of lime use in prehistoric plasters. They occur usually in the form of light brown, low relief mixtures of ill-crystallized portlandite and calcite that also have a hazy, pixilated, white-gray appearance under cross-polarized light. They also occur as high relief cryptocrystalline calcite with moderate, low-order bronze brown birefringence colors. Lime lumps are readily defined from their transitional textures, shrinkage fractures, and occasionally colloidal forms and reaction rims with the surrounding matrix. Binding matrix, however, is the only proof of intentional use of lime, because burnt limestone fragments could have been accidentally added in the aggregate. Reacted cryptocrystalline or fine crystalline calcitic groundmass, with or without transitional textures, is also characteristic of lime plasters. Good knowledge of the probable sources of raw material for the production of quicklime prevents misidentification of lime, because it constrains the range of possible overlapping textures between natural sediments and lime.

It seems that lime can be easily produced in a few hours from heating porous, soft calcareous materials in temperatures no higher than 800°C. This kind of material was frequently used in the Greek prehistory where the use of lime, even in very small amounts, was widespread. Plaster and mortar were made by mixing damp anthropogenic dirt as aggregate and fragments of quicklime, a technique known as “hot mixing.”

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