

Chemistry of 19th Century Lime Mortar on a *tabique Pampango* (Wattle-and-Daub) from the Philippines

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Abstract: The chemical characterization of a thick lime mortar coating on a wall construction method in the Philippines called *tabique Pampango*, or wattle-and-daub method, is reported in this study. The daub material from a 19th century Spanish Colonial Period church convent in Loboc, Bohol, was analyzed using X-ray fluorescence (XRF), X-ray diffraction (XRD), Fourier Transform Infrared Spectroscopy (FTIR), Thermogravimetric Analysis (TGA) and sieve analysis. These analyses demonstrate that the daub material comprises a largely calcitic binder with clay minerals naturally present in the raw material or clayey soils added intentionally. The presence of clay minerals made the binder hydraulic or hardened through added water. Fine-sized aggregates are abundant in the daub material, making it suitable for non-load bearing applications. This study provided a chemical basis on the construction techniques employed during the colonial period in the Philippines that is useful for advancing conservation work and local historical knowledge.

Keywords: wattle-and-daub, tabique Pampango, XRF, XRD, FTIR, TGA, sieve analysis, Philippines

Química del mortero de cal del siglo XIX en una *tabique Pampango* (Wattle-and-Daub) de Filipinas

Resumen: En este estudio se informa sobre la caracterización química de un revestimiento de mortero de cal espeso en un método de construccíon de muros en Filipinas Ilmado *tabique Pampango*, o método de acacia y barro. El material de embadurnamiento de un convento de la iglesia del período colonial Español del siglo XIX en Loboc, Bohol, se analizó mediante flourescencia de rayos X (XRF), difracción de rayos X (XRD), espectroscopía infrarroja por transformada de Fourier (FTIR), análisis termogravimétrico (TGA) y analisis granulometrico. Estos análisis demuestran que el material de embadurnamiento comprende un aglutinante principalmente calcítico con minerales arcillosos presentes de forma natural en la materia prima o suelos arcillosos añadidos intencionalmente. La presencia de minerales arcillosos hizo que el aglutinante fuera hidráulico o se endureciera mediante la adición de agua. Los agregados de tamaño fino abundan en el material de embadurnamiento, lo que lo hace adecuado para aplicaciones que no soportan carga. Este estudio proporcionó una base química sobre las técnicas de construcción empleadas durante el período colonial en Filipinas que es útil para promover el trabajo de conservación y el conocimiento histórico local.

Palabras clave: accacia y barro, tabique Pampango, XRF, XRD, FTIR, TGA, análisis de tamices, Filipinas

Química da argamassa de cal do século XIX num Tabique Pampango (Wattle-and-Daub) das Filipinas

Resumo: Neste estudo é relatada a caracterização química de um revestimento de argamassa de cal espessa num método de construção de paredes nas Filipinas, denominado tabique Pampango, ou método wattle-and-daub. O material de argamassa de um convento da igreja do período colonial Espanhol do século XIX em Loboc, Bohol, foi analisado mediante Fluorescência de raios X (XRF), Difração de raios X (XRD), Espectroscopia de infravermelho com transformada de Fourier (FTIR), análise termogravimétrica (TGA) e análise de peneira. Estas análises demonstram que o material é composto por um aglutinante amplamente calcítico com minerais argilosos presentes de forma natural na na matéria-prima ou solos argilosos adicionados intencionalmente. A presença de minerais argilosos tornou o aglutinante hidráulico ou endurecido por meio da adição de água. Os agregados de tamanho fino são abundantes, tornando-o adequado para aplicações que não suportam carga. Este estudo proporcionou uma base química sobre as técnicas de construção empregues durante o período colonial nas Filipinas, que é útil para o avanço do trabalho de conservação e para o conhecimento histórico local.

Palavras-chave: argamassa, wattle-and-daub, tabique Pampango, XRF, XRD, FTIR, TGA, análise de peneira, Filipinas

Introduction

The wattle-and-daub construction method is a common building material that has existed since ancient times and is characterized by its various forms and manufacturing traditions depending on the country of origin (Chetwin 2007; Singh 2008; Wyżgoł and Deptuła 2020). It is a combination of two indigenous building materials usually employed for wall panels and sometimes in floorings (Sulas *et al.* 2019). It is composed of the "wattle," which is a woven latticework of wooden strips or wooden slats and plastered with a sticky material called the "daub." The daub can be made of lime mortar, clay, or mud and may be mixed with straw and animal dung (Graham 2003). The central feature of this method is the efficient usage of available local raw materials from the surroundings.

In the Philippines, this method is locally known as *"tabique Pampango"* and is the preferred technique for wall partitions during the Spanish Colonial Period. The term *"tabique"* is Spanish for "partition," and *"Pampango"* is derived from the Philippine province of "Pampanga," where this technique was believed to have flourished. This version of wattle-and-daub is described as a thin wall made from a scaffolding of interwoven pieces of bamboo or other wood material and patched with a mixture of lime and sand. The daub serves to achieve a smooth outer finish and makes the wattle sturdier (Jose 1991; Jose 2003). Only few structures in the Philippines that employed this technique have survived and are mostly found in wall partitions of old church convents and clerestory in the provinces of Bohol, Cebu, and Siquijor (Sembrano 2016).

The chemical composition of the daub is the focus of this research due to its susceptibility to variations caused by human interventions during production and chemical changes involved in the complex raw material. In the Philippines, it is believed that lime mortar technology was introduced during the Spanish Colonial Period and influenced how daubs are locally produced. The scarcity of chemical information on daubs made during the 19th century representing a combination of indigenous knowledge with colonial architectural influence is what this study aims to address. Chemical studies on the daub material in the wattle composite are mostly related to archaeological contexts. Different analytical techniques were utilized on excavated daub samples to determine the firing temperature during production (Parr and Boyd 2002; Boyd and Parr 2007), the manufacturing techniques employed based on the composition (Amicone et al. 2020; Ceccarelli et al. 2020), its provenance (Taubald and Biró 2007), and its raw material similarities with other excavated ceramic vessels (Shergur et al. 2003; Duwe 2005). In Portugal, where the daub material is also referred to as tabique, physical and chemical characterizations were performed on several traditional daub structures in the Alto Douro region (Pinto et al. 2010a; Pinto et al. 2010b). This region is known to be abundant in *tabique* technology and the study has provided baseline information for

encouraging earth-based methods for future construction (Cardoso and Pinto 2015). It is evident from these examples that more chemistry research needs to be pursued for this material specially for establishing the different regional diversities.

This study described the daub material in the tabique Pampango method adorning the inner walls of the mid-19th century church convent at the Municipality of Loboc in Bohol, Philippines. The Catholic mission in Loboc is one of the oldest coordinating centers for the entire province of Bohol, established by the Society of Jesus or the Jesuits in the early 17th century. The church constructed in coral stone traces its origin in the early 18th century, and the convent, where the sample was obtained, is attached at its rear end. Loboc has the only convent structure built in a three-story configuration in the country. It is constructed in the vernacular "bahay-na-bato" (house of stone) type of architecture, where the lower level is made of coral stones, and the upper level is made of wood (Javellana 1998; Jose 2001). The convent, together with the church, was severely damaged by an earthquake in 2013 (Aning and Uy 2013). The daub sample was subjected to X-ray fluorescence (XRF), X-ray diffraction (XRD), and Fourier transform infrared (FTIR) spectroscopy for detailed chemical and mineralogical analysis. Information on the hydraulic nature of the binder and other thermal transformations are established using the thermogravimetric analysis (TGA). Sieve analysis was also employed to know the distribution of particle sizes in the daub. The result of this study contributes to the understanding of the material origin and manufacturing process of daub, especially its local adoption in a humid and tropical environment like the Philippines. This study is also the first scientific report on the chemical analysis of daub in a tabique Pampango method in the country. Furthermore, future conservation and restoration work on daub materials will benefit from the baseline chemical data provided by this study.

Methodology

—Sample collection

Figure 1 shows the location of the Municipality of Loboc in Bohol, Philippines, where the *tabique Pampango* sample for this study was obtained. The daub sample was gently detached from a wall partition found inside the 2nd floor of the 19th century Loboc church convent in April 2017 [Figure 2]. Some sections of this wall partition were partially damaged by a strong earthquake in October 2013 which facilitated the dislodging of the daub sample by hand. The daub lime is attached to a wattle made of molave (*Vitex parviflora*) horizontal planks. The sample used in this study measured approximately 3.00 in x 1.80 in x 0.60 in. Since it was originally plastered by hand, the thickness of the sample is non-uniform in some portions [Figure 3]. The exposed outer surface of the sample was discarded to prevent contamination during chemical analysis.



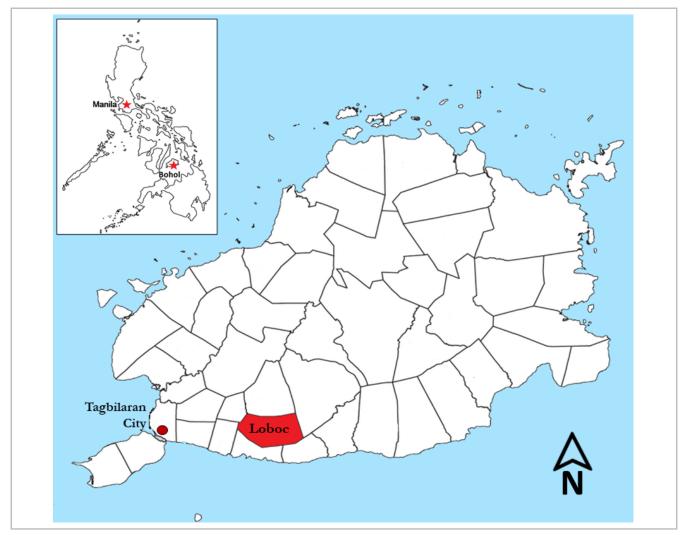


Figure 1.- Location of the Municipality of Loboc in the province of Bohol, Philippines. The capital city of Bohol, which is Tagbilaran City, is also shown in the figure for reference.



Figure 2.- Wall partition using the *tabique Pampango* (wattle-and-daub) technique. (A) and (B) shows the daub collection site with the exposed wattle frame. (C) is the view of the wall partition inside the 19th century Loboc church convent

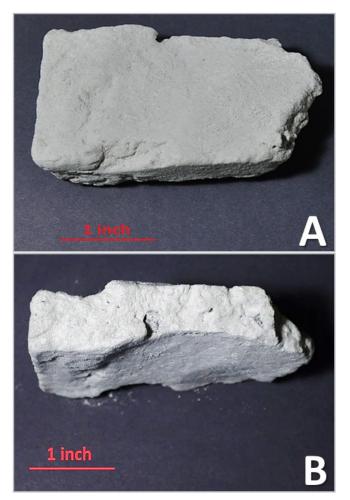


Figure 3.- *Tabique Pampango* mortar sample used in this study. (A) lateral surface view, and (B) view from the side.

Sieve analysis

A portion of the daub sample (15.43g) was gently crushed and carefully disaggregated using a ceramic mortar and wooden pestle to expose the aggregates from the binder fraction. It was done to avoid damaging the attached aggregates such, as broken shells, potteries fragments, or coral chunks if present in the sample. The crushed materials were passed through a series of sieves having the following sizes in accordance with the USA Standard Testing Sieve ASTME11 Specification: 4.75, 2.36, 1.18, 0.600, 0.425, 0.250, 0.150, and 0.075 mm, respectively. For better separation, the whole sieve set-up was placed on a mechanical shaker (US Tyler brand) for 5 min. The fractions that settled in the "pan" were also included in the sieve analysis. It will represent sieve sizes smaller than 0.075 mm, which usually contains the lime binder and clay minerals. Each sieved fractions were weighed carefully to determine the physical distribution of particle sizes in the daub sample.

Fourier Transform Infrared Spectroscopy (FTIR) analysis

The bulk daub sample was prepared for FTIR analysis using the KBr pellet method, and the spectrum was collected on a Thermo Scientific Nicolet 6700 FT-IR Spectrophotometer. The KBr (potassium bromide) pellet was prepared by grinding together an approximate ratio of one part daub sample with three parts anhydrous KBr powder and pressed into a thin, translucent film. The pressed pellet was repeatedly scanned in the FTIR 16 times over the mid-infrared region (4000-500 cm⁻¹) at a resolution of 4 cm⁻¹. The spectrum was reported in percent transmittance relative to wavenumber.

Portable X-Ray Fluorescence (pXRF) analysis

An Olympus Vanta C series handheld X-ray fluorescence analyzer provided elemental composition data of the bulk daub sample. The excitation source is a rhodium (Rh) anode (4-watt, 8-40 kV) with a silicon drift detector. The sampling area was set to 10 mm, and the acquisition followed the Geochem software mode. Total testing time was 45 seconds (Beam 1: 15 sec.; Beam 2: 30 sec.). A raw limestone obtained in Tagbilaran City, capital of Bohol Province, was also sampled using the same parameters for comparison with the daub sample. Results are reported in single element composition, and undetected elements such as carbon and oxygen are classified under the loss on ignition (LOI).

X-ray Diffraction (XRD) analysis

Information on the mineral content in the bulk daub sample was determined in an Olympus TERRA-248 InXitu portable X-ray Diffractometer. The X-ray radiation source used to analyze the sample originated from a Co-K α peak (λ = 1.7903 Å). After homogenization, The XRD data were collected in the °2 θ range of 3.00 to 55.00 with 2 seconds measuring time per step. The mineral phases were interpreted using MATCH! Phase Identification from Powder Diffraction software developed by Crystal Impact (Version 3.7.1.132 downloaded February 12, 2019) and verified through the Mineralogy Database created by David Barthelmy at www.webmineral.com.

Thermogravimetric Analyzer (TGA) analysis

Thermal analysis on the bulk daub sample was carried out on a TA Instruments Discovery TGA55. The sample was pulverized and carefully placed in a small platinum pan (Platinum HT) for high-temperature heating. TGA recorded an initial sample weight of 9.973 mg. Temperature was increased from ambient room temperature (~22°C) to 1,000°C at a ramping rate of 10°C/minute in a nitrogen gas environment. The percentage weight loss was plotted together with the temperature change and the 1st derivative plot was also determined.

Results and Discussion

—Particle distribution

Employing sieve analysis provided information on the particle size distribution of the *tabique Pampango* daub

sample. The sieved data was interpreted based on the soil classification system of the American Society for Testing and Materials (ASTM) (Holtz and Kovacs 1981). The grain size distribution [Figure 4] has the following ranges: coarse sand (4.75 to 2.00 mm), medium sand (2.00 to 0.425 mm), fine sand (0.425 to 0.075 mm) and silt (< 0.075 mm). The weight distribution is highest at the pan (22.75%), showing that the daub sample is mainly classified as silt. Materials retained from 0.075 mm sieve size to lower sizes (i.e., pan) are usually associated with lime or CaCO₃ and minor clay minerals. This observation was followed by the sieve size at 0.250 mm (20.61%) and classified as fine-sized aggregates. The absence of coarse-sized aggregates is also evident from the sieve size at 4.750 mm and the minimal amount retained at 2.360 mm sieve size (0.52%). These results show that the daub sample is non-load bearing or not intended for supporting heavy loads (Cayme et al. 2018). Results of the distribution graph provided an idea of the manufacturing process that involves the use of a substantial amount of lime binder and finer-sized aggregates. Crush seashells or broken potteries are also not observed from the particle distributions in contrast to a lime mortar reported from a 19th century church ruin from Manila, Philippines (Cayme and Asor Jr 2016). Hence, preparation methods would differ depending on the available raw material.

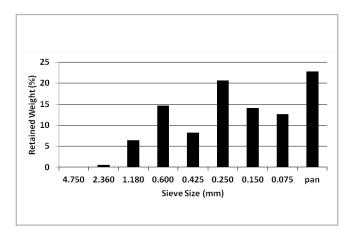


Figure 4.- Percentage weight distribution of the various sieved fractions in the *tabique Pampango* daub mortar sample.

The logarithmic plot [Figure 5] shows the percent finer (% finer) or the cumulative percentage of the daub sample passing through a given mesh size. The grain size diameters (D) passing through the sieve by weight at 10% (D10, 0.0257), 30% (D30, 0.125) and 60% (D60, 0.325), respectively, were extrapolated from the plot. The coefficient of uniformity (Cu, 12.65) and the coefficient of curvature (Cc, 1.87) was computed from these D values. A Cu value greater than four and a Cc value between 1 to 3, which the daub sample is categorized, are considered well-graded (Cayme and Asor Jr 2016). Even though some sieve sizes are not present in the sample, such as the 4.750 mm size, in general, it is still considered to be well represented, and the manufacturer has knowledge on the amount of binder and aggregates to be combined.



Figure 5.- Grading curve distribution of the *tabique Pampango* daub mortar sample.

Elemental composition

The tabique Pampango daub sample is primarily composed of calcium (38.68%), silicon (4.25%), and aluminum (3.38%), respectively [Table 1]. These elements can be traced to the raw materials used for its production, which are the lime binder, mostly CaCO₃, and sand or soil aggregates, largely silicates (SiO₂) with aluminates (Al₂O₃) (Cayme and Asor Jr 2017). A comparison with the elemental composition of an unfired raw limestone obtained from Bohol shows the same trend in abundance of major elements (i.e. Ca, Si, and Al). This observation indicates that the raw limestone is not entirely made of CaCO, but instead argillaceous in nature. However, the amount of silicon and aluminum in the daub sample is significantly higher than the raw limestone, which implies that additional sand or soil are mixed together with the lime binder, as supported by the sieve analysis. The amount of calcium was also found to be different from the raw limestone. These changes are attributed to the differences in processing the starting raw materials, wherein, during the olden times, strict standards in production are not yet in place. The sieve analysis results also yielded a significant amount of fine to coarse-sized aggregates added to the binder, confirming the variations in composition. Furthermore, the amount of iron is almost 50% higher in the daub sample, implying that clayey-type minerals were fired together with the sample during the calcination of lime (Cayme 2021a).

Magnesium is not present in the daub sample, indicating that the carbonates are calcitic in origin. This absence is also consistent with the raw limestone, affirming that the lime may have come from within the Bohol island. The LOI value of about 52.0% is attributed to organic contaminants during the calcination of lime and other hydrated compounds (Kang *et al.* 2019). During the colonial period, one local method of firing lime is to sandwich it in between bamboo slats and burned openly (Reyes 1928). The charred organic substance can mix with the lime causing these impurities.

Binder type is estimated by computing the cementation index (CI) using Equation 1. From the CI value, the binder can be classified as hydraulic (>0.7), moderately hydraulic (0.5 to 0.7), low hydraulic (0.3 to 0.5), and aerial or non-

hydraulic (<0.3), respectively. Hydraulic behavior is the ability of the plaster to harden with the addition of water. This behavior is made possible by mixing pozzolans, where a typical example is clay. A non-hydraulic behavior means that the binder will recarbonate entirely through reaction with air. Based on the computed value, the CI of the daub sample is 0.41 and classified as having low hydraulicity (Gleize *et al.* 2009). Hence, clay may have been involved in the production process.

[1]
$$CI = \frac{(2.8) (\% SiO_2) + (1.1) (\% Al_2O_3) + (0.7) (\% Fe_2O_3)}{\% CaO + (1.4) (\% MgO)}$$

	<i>tabique Pampango</i> daub sample	Raw limestone Percentage Amount (%) 46.69	
Element	Percentage Amount (%)		
Ca	38.69		
Si	4.25	2.88	
AI	3.38	3.09	
Fe	0.457	0.290	
S	0.361	0.014	
Sr	0.359	0.021	
Р	0.221	-	
К	0.211	-	
Ti	0.047	0.067	
Mn	0.011	0.015	
Zn	0.011	0.004	
Zr	0.007	-	
Th	0.003	-	
Cu	0.002	0.002	
Ni	-	0.002	
LOI*	51.99	46.92	

* LOI - loss on ignition

Table 1.- Elemental composition of the daub mortar samplecompared to a raw limestone using the portable X-ray Fluorescence(XRF).

Mineralogical characterization

The principal mineral phases that usually occur in historic lime mortar samples applicable to the daub samples in this study are recarbonated $CaCO_3$ or calcite and silicates. Calcite is the main mineral in the lime binder, and the silicates represent the added aggregates (Cayme and Asor Jr 2017). Possible clay minerals found naturally in the raw material or added intentionally during the production process may have been present due to the variable amount of aluminum, silicon, and iron detected in the EDXRF. The carbonate-containing mineral, calcite, composed mainly of $CaCO_3$, has the most distinct peaks in the FTIR spectrum, as illustrated in Figure 6. The noticeable vibrations associated with the carbonate ion, CO_3^{-2} , are the intense broad peak

centered at 1429 cm⁻¹ assigned to the C-O asymmetric stretching (v3), and the two sharp peaks at 712 cm⁻¹ and 875 cm⁻¹, attributed to C-O in-plane bending (v4) and C-O out-of-plane bending (v2) vibrations, respectively. Minor absorption peaks for calcite includes the C-O combination modes at 1797 cm⁻¹ (v1 + v4) and 2515 cm⁻¹ (2v2 + v4). Overtones at 2872 cm⁻¹ and v3 combination bands at 2966 cm⁻¹ also add to the identification of calcite. The absorption peak at 712 cm⁻¹ clearly shows that the daub sample is mainly calcitic lime rather than dolomitic, which is reported in the literature to have an absorption peak at 727cm⁻¹ (Müller *et al.* 2014; Brunello *et al.* 2019).

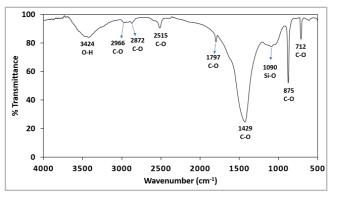


Figure 6.- Fourier Transform Infrared Spectroscopy (FTIR) spectra of the *tabique Pampango* daub mortar sample.

The XRD diffractogram [Figure 7] further supports the identification of calcite through the high-intensity peak at 34.30 (2 θ), 3.033 Å. Other d-spacing values for calcite, readily observed from the diffractogram, includes 3.846 Å, 2.496 Å, 2.284 Å, and 2.095 Å, respectively. The absence of dolomite phases from the XRD also supports the FTIR and EDXRF data. Hence, the raw material used for the lime binder in the *tabique Pampango* is calcium-rich and originated from recarbonated CaCO₃. Two possibilities for the origin of the lime can be deduced; it may either have been sourced directly from marine shells or quarried limestone rocks initially formed from marine deposits ages ago. The latter is a typical sedimentary rock type common in the Bohol island (Travero 2016).

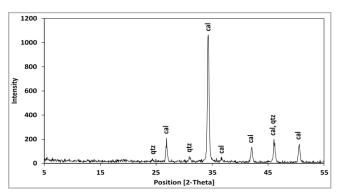


Figure 7.- X-ray powder diffraction (XRD) pattern of the *tabique Pampango* daub mortar sample. Abbreviation: cal – calcite, and qtz – quartz.



For the aggregates, quartz or SiO₂ is the general mineral marker for soil, sand, rocks, or clay because it always contains a significant amount of silicon dioxide or the Si-O bond as part of its backbone. Its presence in the daub sample is displayed through the broad FTIR peak for a Si-O asymmetric stretching vibration centered at 1090 cm⁻¹ [Figure 6] (Cayme and Asor Jr 2017). Due to overlapping peaks with the more dominant CaCO₃ and its lower concentration in the sample, this is the only vibration mode for Si-O assigned with confidence in the FTIR spectrum. As a confirmatory method, crystalline patterns for quartz are also identified from the XRD, as shown by the low-intensity peaks having d-spacing values at 4.241 and 3.342 [Figure 7].

The presence of specific clay phyllosilicate minerals was obscured by the broad FTIR peak centered at 3424 cm⁻¹ assigned to the O-H peak from absorbed moisture. The region from 3700 to 3600 cm⁻¹ are also characteristic FTIR vibrations of clay minerals for inner surface O-H groups; hence overlapping can occur [Figure 6] (Müller et al. 2014). XRD was unable to detect crystalline patterns for clay minerals due to its very low concentration relative to both quartz and CaCO₃. It is not unusual for clay to be part of the soil or sand aggregates added to the daub sample due to Bohol's general clayey type soil composition. Furthermore, FTIR absorption peaks for possible organic additives in the daub were also not detected. Historical documents in the Philippines speculate that albumin from eggs, plant extracts, and molasses is sometimes added to the mortar mixture, but no scientific study has proven this (Jose 2003). Hence, the daub sample in this study was manufactured only from typical lime binder and sand or soil aggregates.

Thermal Analysis

Thermal transformation products resulting from the controlled heating of the *tabique Pampango* daub sample and monitoring the weight change would enable the identification of the binder type used for the sample. Figure 8 shows a continued weight loss from 22°C until it plateaued at around 720°C. Maximum weight loss at about 700°C is typical of recarbonated CaCO₃ and not from pure CaCO₃ (Cayme *et al.* 2021b). Hence, the sample is a processed material. Specific temperature ranges in the thermograph [Table 2] correspond to the processes

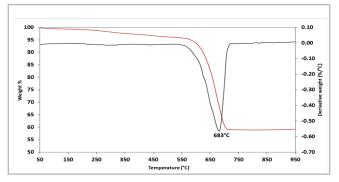


Figure 8.- Thermogravimetric data of the *tabique Pampango* daub mortar sample. The black line corresponds to the 1st derivative plot and the red line is the weight loss.

of dehydration, dehydroxylation, and decomposition (Moropoulou *et al.* 1995a). The dehydration range eliminates the absorbed water (until 120°C) and crystallized hydrated salts from 120°C to 200°C in the sample. The combined weight loss of less than 1.0% in these temperatures is likely from its nature as a wall material directly exposed to the atmosphere, thus, minimizing moisture inclusions.

Dehydrolylated clay minerals that act as pozzolans during the original firing of the binder raw material react with lime (CaCO₃) in the presence of water (hydration) to form hydraulic mineral phases such as calcium-silicate-hydrates and calcium-aluminum-silicate-hydrates. The temperature range between 200°C to 600°C is attributed to the removal of these chemically bound hydraulic water from the sample. The weight loss of 5.38% is typical for hydraulic lime mortars. The decomposition of the CaCO₃ binder by the release of CO₂ occurs at a temperature higher than 600°C (Moropoulou *et al.* 1995b). The maximum weight loss was observed within the approximate range of 600°C to 750°C and centered at 683°C as seen from the 1st derivative plot [Figure 8].

Temperature change (°C)						
T < 120°C	120 - 200°C	200 - 600°C	T > 600°C	Total Weight Loss	CO ₂ /H ₂ O	
0.62 %	0.28%	5.38 %	34.02 %	40.30 %	6.32	

Table 2.- Weight loss in percentage (%) per temperature range ofthe tabique Pampango daub mortar sample.

Furthermore, the early decomposition temperature, starting at about 600°C, also suggests that dehydrolylated clay minerals (e.g., illite) are present during the original firing of the lime raw material. These clay minerals are present in minor amounts (< 1.0%) that the XRD may not have detected but implied from the results of the EDXRF data having typical chemical elements originating from clay. This minor amount of clay is still significant enough to elicit a hydraulic behavior on the daub sample. The level of hydraulicity is confirmed further by obtaining the CO_2/H_2O ratio, which is the weight loss above 600°C (i.e., CO_2), and the hydraulic water within the 200°C to 600°C range. The ratio of 6.32 is within the characteristic range (between 4.0 to 9.0) of hydraulic lime mortars. This behavior is possibly due to clayey soils mixed with the lime binder during its production (Moropoulou *et al.* 2005).

Conclusions

This study provides a foundation for future chemical analysis of a 19th century *tabique Pampango* in the Philippines. It will further motivate the scientific discussions of wattle-anddaub methods in the Southeast Asian region and highlight the influence of Spanish building methods on its former overseas territories. The daub sample from the Loboc convent yielded significant amounts of calcite and quartz, which is expected for a mortar made of lime binder and sand aggregates. Clay minerals added to the binder have made the daub hydraulic, which hastens the hardening of the material instead of entirely carbonating on exposure to atmospheric carbon dioxide. This interpretation represents an advanced way of conducting the wattle-and-daub method due to the added pozzolan materials in the binder. The results also suggest no difference between the lime mortar used for load-bearing structures compared to the daub material, except for the sizes of aggregates. It shows that the binders are prepared the same and will only differ on the type of aggregates depending on the structure's purpose. The daub sampling sites should be increased in future studies to determine the variations across different provinces in the country and, broadly, to other former territories of the Spanish Empire.

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References

AMICONE, S., CROCE, E., CATELLANO, L., VEZZOLI, G. (2020). "Building Forcello: Etruscan wattle-and -daub technique in the Po Plain (Bagnolo San Vito, Mantua, Northern Italy", *Archaeometry*, 62(3):521-537. <u>https:// doi.org/10.1111/arcm.12535.</u>

ANING, J., UY, J.R. (2013). "Destruction of heritage churches lamented". Philippine Daily Inquirer <u>https://newsinfo.inquirer.net/507871/</u> <u>destruction-of-heritage-churches-lamented</u>, [accessed: 3/30/2021].

BOYD, W.E., PARR, J.F. (2007). "XXIV. Geochemical analysis of fragments of burnt and impressed clay (daub) from the archaeological layers at Noen U-Loke, and the determination of its probable origin". In: *The origins of the civilizations of Angkor: The excavation of Noen U-Loke and Non Muang Kao* (Vol. II), Higham, C.F.W., Kijngam, A., Talbot, S. (eds.). Bangkok: The Thai Fine Arts Department, 465-474.

BRUNELLO, V., CORTI, C., SANSONETTI, A., TEDESCHI, C., RAMPAZZI, L. (2019). "Non-invasive FTIR study of mortar model samples: Comparison among innovative and traditional techniques", *The European Physical Journal Plus*, 134:270. https://doi.org/10.1140/epjp/i2019-12667-1.

CARDOSO, R., PINTO, J. (2015). "Tabique walls composite earth based material characterization in the Alto Douro wine region, Portugal", *Construcții Journal*, 15(2):3-11.

CAYME, J.M., ASOR J.R., A. (2016). "Characterization of historical lime mortar from the Spanish colonial period in the Philippines", *Conservation Science in Cultural Heritage*, 16:33-44. <u>https://doi.org/10.6092/issn.1973-9494/7164</u>

CAYME, J.M., ASOR J.R., A. (2017). "Calcium content of lime mortars from 19th century church ruins in the Philippines using volumetric analysis", *Malaysian Journal of Analytical Sciences*, 21(5):1080-1090. <u>https://doi.org/10.17576/mjas-2017-2105-10</u>.

CAYME, J.M., AURELLANO, R.M., CABRAL, C.L., ALONZO, G.A., ASOR JR., A. (2018). "Assessing the composition of 19th century lime mortars from a mission chapel in the former Hacienda de San Isidro de Mariquina Philippines", *Jurnal Kimia Sains dan Aplikasi* (Journal of Scientific and Applied Chemistry), 21(3):131-138. <u>https://doi.org/10.14710/jksa.21.3.131-138</u>.

CAYME, J.M. (2021a). "Analytical chemistry methods of estimating the original firing temperature of bricks from a 19th century convent in the Philippines: Perspective of a Southeast Asian country", *SPAFA Journal*, 5. <u>https://doi.org/10.26721/spafajournal.2021.5.651</u>.

CAYME, J.M., BERMEJO III, A., FRANCIA, C.A.E., ASOR JR., A., MIRANDA, E. (2021b). "Spanish Colonial Period bricks from churches in Laguna, Philippines: A preliminary chemical characterisation using X-ray diffraction, energy dispersive X-ray flourescence and Fourier transform infrared", *Journal of Physical Science*, 32(2):105-124. <u>https://doi.org/10.21315/jps2021.32.28</u>.

CECCARELLI, L., MOLETTI, C., BELLOTTO, M., DOTELLI, G., STODDART, S. (2020). "Compositional characterization of Etruscan earthen architecture and ceramic production", *Archaeometry*, 62(6):1130-1144. https://doi.org/10.1111/arcm.12582.

CHETWIN, J.N. (2007). "XXIII. The remains of wattle and daub structures". In: *The origins of the civilizations of Angkor: The excavation of Noen U-Loke and Non Muang Kao* (Vol. II), Higham, C.F.W., Kijngam, A., Talbot, S. (eds.). Bangkok: The Thai Fine Arts Department, 447-464.

DUWE, S. (2005). "Pots, daub, and neutrons: A pilot compositional analysis of Hungarian Early Copper Age clay assemblages", *La Tinaja: A Newsletter of Archaeological Ceramics*, 16(1-2): 2-5.

GRAHAM, T. (2003). "Wattle and daub: Craft, conservation and Wiltshire case study". Master of Science in the Conservation of Historical Buildings at the University of Bath. Dept of Architecture and Civil Engineering. https://doi.org/10.13140/2.1.1875.2806. [accessed: 3/30/2021].

GLEIZE, P.J.P., MOTTA, E.V., SILVA, D.A., ROMAN, H.R. (2009). "Characterization of Historical Mortars from Santa Catarina (Brazil)", *Cement and Concrete Composites*, 31(5): 342-346. <u>https//doi.org/10.1016/j.cemconcomp.2009.02.013</u>.

HOLTZ, R., KOVACS, W. (1981). *An Introduction to Geotechnical Engineering*. New Jersey: Prentice-Hall, Inc.

JAVELLANA, R. (1988). "Angels and Gargoyles of Loboc Church", *Philippine Studies*, 36(1): 88-97.

JOSE, R.T. (1991). *Simbahan: Church Art in Colonial Philippines 1565-1898*. Makati: Ayala Museum.

JOSE, R.T. (2001). *Visita Iglesia Bohol: A Guide to Historic Churches*. Manila: National Commission for Culture and the Arts (NCCA).



JOSE, R.T. (2003). "Palitada: Skin of the church". In: Skin Surface Essence. Manila: Ateneo Art Gallery, Ayala Foundation, Inc., Eugenio Lopez Foundation, Inc., and Museum Pambata Foundation, Inc., 13-29.

KANG, S-H., LEE, S-O., HONG, S-G., KWON, Y-H., (2019). "Historical and scientific investigations into the use of hydraulic lime in Korea and preventive conservation of historic masonry structures", *Sustainability*, 11: 5169. <u>https://doi.org/10.3390/su11195169</u>.

MOROPOULOU, A., BAKOLAS, A., MICHAILIDIS, P., CHRONOPOULOS, M., SPANOS, CH. (1995a). "Traditional technologies in Crete providing mortars with effective mechanical properties". In: *Structural Studies, Repairs and Maintenance of Historical Building. WIT Transactions on The Built* Environment, Brebbia, C.A. (ed.). WIT Press, 15:151-161. <u>https://</u> doi.org/10.2495/STR950171.

MOROPOULOU, A., BAKOLAS, A., BISBIKOU, K. (1995b). "Characterization of ancient, byzantine and later historic mortars by thermal and X-ray diffraction techniques", *Thermochimica Acta*, 269/270: 779-795. <u>https://doi.org/10.1016/0040-6031(95)02571-5</u>.

MOROPOULOU, A., BAKOLAS, A., ANAGNOSTOPOULOU, S. (2005). "Composite materials in ancient structures", *Cement & Concrete Composites*, 27: 295-300. <u>https://doi.org/10.1016/j.cemconcomp.2004.02.018</u>

MÜLLER, C.M., PEJCIC, B., ESTEBAN, L., PIANE, C.D., RAVEN, M., MIZAIKOFF, B. (2014). "Infrared attenuated total reflectance spectroscopy: An innovative strategy for analyzing mineral components in energy relevant systems", *Scientific Reports*, 4:6764. <u>https://doi.org/10.1038/srep06764</u>.

PARR, J.F., BOYD, W.E. (2002). "The probable industrial origin of archaeological daub at an Iron Age site in Northern Thailand", *Geoarcaheology: An International Journal*, 17(3): 285-303. <u>https://doi.org/10.1002/gea.10013</u>.

PINTO, J., VARUM, H., CRUZ, D., SOUSA, D., MORAIS, P., TAVERES, P., LOUSADA, J., SILVA, P., VIEIRA, J. (2010a). "Characterization of traditional tabique constructions in Douro North Valley region", *WSEAS Transactions on Environment and Development*, 2(6): 93-102.

PINTO, J., VARUM, H., CEPEDA, A., TAVERES, P., LOUSADA, J., SILVA, P., VIEIRA, J. (2010b). "Study of the traditional tabique constructions in Alto Tâmega region". In: *The Sustainable World. WIT Transactions on Ecology and the Environment*, Brebbia, C.A. (ed.). WIT Press, 142:299-307. https://doi.org/10.2495/SW100281.

REYES, F.D. (1928). "The lime industry of the Philippine Islands". *The Philippine Journal of Science*, 36(2): 139-155.

SEMBRANO, E.A.M. (2016). "Siquijor's healing sights: Churches made of ancient 'tabique pampango'", *Philippine Daily Inquirer*. <u>https://lifestyle.inquirer.net/243860/siquijors-healing-sights-churches-made-of-ancient-tabique-pampango/</u>. [accessed: 3/30/2021].

SHERGUR, J., POPELKA, R.S., ROBERTSON, J.D., POLLACK, D. (2003). "Distinct chemical patterns in late Mississippian Caborn-Welborn ceramics of the lower Ohio river valley", *North American Archaeologist*, 24(3): 221-243. <u>https://doi.org/10.2190/PT2Q-AV4X-UAWX-EJ1A</u>. SINGH, U. (2008). A history of ancient and early medieval India: From the stone age to the 12th century. India: Dorling Kindersley Pvt. Ltd.

SULAS, F., KRISTIANSEN, S., WYNNE-JONES, S. (2019). "Soil geochemistry, phytoliths and artefacts from an early Swahili daub house, Unguja Ukuu, Zanzibar", *Journal of Archaeological Science*, 103: 32-45. <u>https://doi.org/10.1016/j.jas.2019.01.010</u>.

TAUBALD, H., BIRÓ, K.T. (2007). "Archaeometrical analysis of Neolithic pottery and comparison to potential sources of raw materials in the immediate environment of the settlements – A first summary of the MÖB-DAAD exchange program", *Archeometriai Műhely*, 2.

TRAVERO, J.T. (2016). "Soil types and geographical forms of the degraded uplands of Bohol, Philippines", *International Journal of Environmental and Rural Development*, 7(2): 1-5. <u>https://doi.org/10.32115/ijerd.7.2_1</u>.

WYŻGOŁ, M., DEPTUŁA, A. (2020). "Living outside the wall: The quarter of wattle-and-daub houses in old Dongola of the Funj Period", *Journal of African Archaeology*, 18: 109-132. <u>https://doi.org/10.1163/21915784-20200005</u>.

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