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An Archaeometric Investigation of the White-infilled Decorations in Għar Dalam Phase Pottery

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ABSTRACT

The study explores complementary archaeometric analyses applied to Għar Dalam phase white-infilled decorations (*c.* 6,000-5,400 BC), traditionally believed to be composed of gypsum as proposed by Trump (1966). This research represents the first multi-sample and multi-site study, examining a total of five samples from Skorba and Santa Verna. Using optical microscopy and SEM for microstructure observation, EDS for chemical composition and μ-XRD for mineralogical data, the analyses revealed that calcite was the primary decorating material, consistently found alongside quartz, with additional minerals including dolomite, aragonite and fluorapatite. No gypsum was detected, challenging the previously held hypothesis. Furthermore, the study underscores the technological knowledge of the potters, reflected in their selection of locally available materials and their understanding of factors in the *chaîne opératoire* including material procurement, binder use and firing processes.

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Chapter 1: Introduction

The Għar Dalam phase, possibly dating back to 6,000-5,400 BC (McLaughlin *et al* 2020a: 38), represents the earliest known period of human habitation in the Maltese Islands. It was during this period that distinctive Għar Dalam pottery emerged, displaying similarities with the Stentinello pottery industry (5,600-4000 cal BC) (Freund *et al* 2015: 208), named after its type site in Sicily (Morter 2010: 9). Importantly, both pottery traditions share common decorative features, including the use of a white infill for their impressed decorations. These connections between the pottery traditions of Għar Dalam and Stentinello highlight cultural exchanges and potential influences between the Maltese Islands and Sicily during the Neolithic period (Sagona 2015: 23), underscoring the significance of investigating the composition, provenance and technology of pottery and decorations, including the white infills, from this period in both Malta and Sicily.

The tradition of applying white infills to impressed pottery decorations dates back to the earliest Neolithic phases in Europe. Archaeological discoveries, such as those at Tell Samovodene in Bulgaria (5750-5400 BC) (Gencheva 1992: 639) and within the Vinča culture (5500-4500 BC) (Perišić *et al* 2016: 1415), attest to its early practice. This tradition persisted through subsequent historical periods, evidenced by examples in Transdanubian Encrusted Pottery (2000-1500 BC) (Sofaer and Roberts 2016: 480) and pottery from the Zhabotyn settlement in Ukraine (8th century BC) (Kulkova *et al* 2020: 931).

Archaeometric analyses conducted on white infills from sherds found across Europe has revealed a variety of distinct mineral compositions (Odriozola and Perez 2007, Perišić *et al* 2016, Sofaer and Roberts 2016, Kulkova *et al* 2020, Pirovska *et al* 2020, Opriş *et al* 2022). These compositions primarily consisted of either kaolinite, gypsum, hydroxyapatite, lime or calcite, with calcite and hydroxyapatite being the most frequently identified.

Scientific analysis of the white infill in Għar Dalam phase pottery has not been published, leaving room for hypotheses regarding its composition. Emmanuel Magri was among the earliest to document the presence of white infill in Maltese pottery decorations, referring to it as "chalk-inlaying" and describing it as "white carbonate of lime" (1906: 6). Despott (1923: 24) provides one of the earliest references to the white-infilled decorations of Għar Dalam phase pottery, while Trump was among the first to propose a hypothesis about the composition(s) of the white infill, suggesting that it could be derived from gypsum, a material

found in local clay (1966: 23). Subsequent mentions in the literature, which similarly lacked scientific analyses of the white infill, also reiterated this hypothesis (Evans 1971: 209, Guzzardi 2008: 41, Veen and van der Bloom 1992: 45, Sagona 2015: 30, Malone *et al* 2020a: 325). This study aims to assess the validity of these hypotheses by investigating the potential mineral compositions that could have been employed in producing the white-infilled decorations in Għar Dalam phase pottery. This research project complements and expands upon the MaltaPot project (https://www.um.edu.mt/projects/maltapot/), which included the same pottery sherds used in this study, among others, in its scope. This project aimed to investigate Early Neolithic pottery in the Maltese Islands, focusing on fabric classification, material composition for provenance studies, and manufacturing techniques to understand where and how these vessels were made.

Understanding the composition and characteristics of the white-infilled decorations in Għar Dalam phase pottery is relevant to Maltese archaeology. Analysing the white infill contributes to the understanding of pottery decoration techniques and technology during that era. By ascertaining the composition of the infill, insights can be gained into the materials and techniques employed by the craftspeople/potters. Secondly, examining the white-infilled decorations aids in evaluating the homogeneity (or otherwise) of practices in the Għar Dalam phase. Homogeneity in using specific materials and techniques across various white infills could imply shared craft practices within the Għar Dalam phase. However, it could also be due to the types of resources available, making the study of white infills an opportunity to investigate resource use. Lastly, this research project will contribute to the field by providing comparative data for future studies aiming to compare Għar Dalam phase pottery to other Maltese sites or Stentinello pottery from Sicily and Calabria (Scarcella *et al* 2011).

1.1 Scope

The scope of this research project is to understand the composition of the white infill found in the decorations of five Għar Dalam phase pottery sherds (**Figure 1**).

Figure 1: The five sherds that will be analysed in this study: (a) G1002, (b) G1004, (c) G1005, (d) G1021 and (e) G1030.

1.2 Aims and Objectives

This study will endeavour to achieve the following aims in order to satisfy its scope.

1. The first aim is to document the application and occurrence of the white infill found on Għar Dalam phase pottery from the Maltese Islands.

   The objectives established to accomplish this aim are as follows:

   a) Conduct a visual examination of the white infill to document its appearance, including colour, texture and surface features. The examination will include both naked-eye observations and the use of a magnifying glass or low-power microscope. Documentation will be conducted through the implementation of macroscale and microscale photography.

b) Carry out a microscopic examination of the white infill to analyse its structure at the micro-scale (64x-1.13Kx). The examination will primarily employ a Variable Pressure Scanning Electron Microscope (VP-SEM) to capture images of the inlay's microstructure.

2. The second aim is to analyse the chemical and mineral composition of the white infill of five sherds and compare them.
   The objective established to accomplish this aim is as follows:
   a) Utilise Energy-Dispersive Spectroscopy (EDS) to preliminarily analyse the chemical composition of the white infills and establish a hypothesis regarding their composition.
   b) Employ Micro-X-ray Diffraction (μ-XRD) to further investigate the mineralogical composition of the white infills and confirm or refine the hypothesis established through EDS analysis.

3. The final aim is to suggest possible manufacturing techniques used to produce and apply the white infill within the *chaîne opératoire*.
   The objective established to accomplish this aim is as follows:
   a) Use the results obtained from the analytical techniques and the literature to develop hypotheses about the materials and manufacturing techniques involved in producing and applying the white infill.

Chapter 2: Literature Review

2.1: Għar Dalam Phase

2.1.1 Overview

Introduction of Għar Dalam phase in Malta

The earliest identifiable presence of material culture unearthed on the Maltese Islands can be traced back to the pottery associated with the Early Neolithic Għar Dalam phase (McLaughlin *et al* 2020a: 31). This nomenclature is derived from a cave situated in Wied Dalam, a valley in Birżebbuġa, which yielded a considerable quantity of this style of impressed pottery (McLaughlin *et al* 2020a: 31). The FRAGSUS Project has proposed a new chronological sequence, suggesting that Għar Dalam phase could date as far back as 6000 BC (**Table 1**) (McLaughlin *et al* 2020a: 39), although these early dates of this chronology have not been corroborated with other dating campaigns.

Period	Phase	Trump 2009		*FRAGSUS*	
		Start	**End**	**Start**	**End**
Neolithic	Għar Dalam	5000 BC	4300 BC	6000 BC	5400 BC
	Skorba	4500 BC	4100 BC	5400 BC	4800 BC
	Fifth millennium hiatus	n/a		4800 BC	3800 BC
Temple Period	Żebbuġ	4100 BC	3700 BC	3800 BC	3600 BC
	Mġarr / transitionary phase	3800 BC	3600 BC	3600 BC	3400 BC
	Ġgantija	3600 BC	3100 BC	3400 BC	3100 BC
	Saflieni	3300 BC	3000 BC	3100 BC	2800 BC
	Tarxien	3100 BC	2400 BC	2800 BC	2400 BC
Bronze Age	Thermi	n/a		2400 BC	2200 BC
	Third millennium hiatus	n/a		2200 BC	2000 BC
	Tarxien Cemetery	2000 BC	1500 BC	2000 BC	1700 BC
	Second millennium hiatus	n/a		1700 BC	1500 BC
	Borġ in-Nadur / Baħrija	1500 BC	700 BC	1500 BC	750 BC

Table 1: The revised timeline of Maltese prehistory resulting from the FRAGSUS Project (Malone *et al* 2020d: 476).

The Għar Dalam pottery styles and the people who initially introduced them to the Maltese Islands can be connected to the Stentinello wares discovered in Sicily and Calabria (McLaughlin *et al* 2020a: 31). The current understanding is that the first settlers of Malta arrived from Sicily during the 6th millennium BC, hence the similarities in their material culture. Stentinello wares are considered as a developed form of another early Neolithic impressed ceramic, commonly referred to as Cardial Ware (McLaughlin *et al* 2020a: 31). Such wares are distinguished by punctuated linear designs created by impressing the notched edge

of the *Cardium edule* cockleshell (Sagona 2015: 23). Substantial connections have also been proposed, linking Malta's Għar Dalam phase to locations such as Monte Kronio in Sicily (Tine 1971: 75), Palma di Montechiaro (also within Sicily) and the island of Ognina (Sagona 2015: 26). Additionally, the discovery of Għar Dalam pottery in Vulpiglia suggests a network of trade interactions between the Maltese Islands and Syracuse (Guzzardi 2008: 41), although these have yet to be characterised and their provenance ascertained.

Għar Dalam Phase Pottery

Għar Dalam phase pottery shares commonalities in typology and decoration with Sicilian Stentinello ware, particularly in its prevalence of globular vessel forms (Evans 1971: 208). Both styles exhibit similar decorative patterns, including geometric motifs such as chevrons and sequences of impressed lines (Richard-Trémeau *et al* 2023a: 6). While the use of stamps for repetitive decorations is a characteristic feature of Stentinello ware (Daniel and Evans 1975: 16), there is limited evidence of this technique in Maltese pottery, with a single possible instance published from Santa Verna (Richard-Trémeau *et al* 2023a: 7) and other examples reported in Għajn Abdul (Scarcella 2011: 176-179) which have yet to be published.

Although Għar Dalam phase pottery exhibits similarities in terms of shape and decoration with the Stentinello ware, it is notable that these two pottery types were discovered in distinct archaeological contexts (Vella Gregory 2021). In Sicily, Stentinello ware was often found in ditches along the eastern coastal plain, with unique settlement configurations in other regions (Morter 1992: 19, Vella Gregory 2021) while in Malta, there is no evidence for ditched settlements, and instead, the settlements might have been located within caves (Vella Gregory 2021). The difficulties in identifying Early Neolithic settlements in Malta could also be explained by the reuse of these sites during later periods (Malone *et al* 2020a: 313).

A method for identifying Għar Dalam phase pottery relies on analysing its decorations as it is the tools and production techniques used that distinguish the Impressed Wares (Vella Gregory 2021). One technique widely employed for impressed wares was comb-stamping, executed on semi-wet clay, resulting in a variety of designs (Vella Gregory 2021). For instance, certain impressed pottery fragments display chevron patterns that have been either incised or stamped, forming a continuous zig-zag design or patterns reminiscent of herringbone (Molitor 1988: 30). Occasionally, these patterns are complemented by the addition of lines created through comb-stamping, demonstrating the integration of various tools and techniques (Vella Gregory 2021). Notably, a pottery fragment from the Għar Dalam phase discovered at Mġarr

(Ta' Ħaġrat) exhibits a pair of parallel lines produced using a serrated comb with two teeth, employing the Alternately Pivoting Stamp (APS) method (Vella Gregory 2021). Furthermore, other Għar Dalam pottery fragments feature incised lines instead of comb imprints (Vella Gregory 2021). These lines display a variety of attributes, including parallel layouts, combinations of parallel and angular lines, and arrangements forming incised patterns (Vella Gregory 2021). **Figure 2** shows an Għar Dalam phase sherd featuring different decorations. Reviewing the different types of impressions/incisions is pertinent to this research project, as this study will involve analysing white infills in various decorations, which could potentially identify distinct compositions associated with specific types of impressions/incisions.

Figure 2: Għar Dalam phase sherd, featuring intricate designs including banded incised chevrons, comb-stamping on the rim and incised channels below the rim (Vella Gregory 2021: 4).

Għar Dalam phase pottery displays enhanced decorations achieved through the application of a white paste. However, this material typically does not survive well within the archaeological record of Malta (Sagona 2015: 30). Despott (1923: 24) provides one of the earliest references to the white-infilled decorations of Għar Dalam phase pottery, while Trump was among the first to propose a hypothesis about the composition(s) of the white infill, suggesting that it could be derived from gypsum, a mineral found in local clay (1966: 23). Subsequent mentions in the literature, which were also not based on scientific analyses, agreed with this hypothesis (Evans 1971: 209, Veen and van der Bloom 1992: 45, Sagona 2015: 30). This study seeks to examine the validity of these hypotheses and explore alternative mineral compositions that may have been used to create the white-infilled decorations in Għar Dalam phase pottery.

Għar Dalam Phase Sites

Numerous Early Neolithic sites are present on the Maltese Islands, and most have scatters of Għar Dalam phase material culture, as depicted in **Figure 3**. Almost all these sites lack structural remains except for the site of Skorba, where a possible Għar Dalam phase wall and hut were found (Trump 1966: 10-11). The discovery of Għar Dalam phase pottery often occurs in the form of scattered sherds, posing challenges in terms of contextualisation and interpretation (Richard-Trémeau et al 2023a: 6). Examples include Taċ-Ċawla and Ġgantija in Gozo, as well as Kordin III in Malta (Richard-Trémeau et al 2023a: 6). In the stratigraphic layers where Għar Dalam phase pottery is present, it has been observed that later materials become intermixed (Richard-Trémeau et al 2023a: 6). For example, recent archaeological studies at Santa Verna and Skorba revealed a consistent pattern of finding Għar Dalam phase pottery alongside Skorba phase pottery, and occasionally even pottery from the later Temple period (McLaughlin et al 2020b: 142, 154). To date, no pottery production sites from the Għar Dalam period have been identified in the Maltese Islands, so our knowledge of local pottery production during this time is entirely based on the analysis of pottery sherds (Richard-Trémeau et al 2023a: 6).

Sites

① Birkirkara [GD]
② Borġ in-Nadur [GD]
③ Borġ l-Imramma [GD]
④ Buqana [Z]
⑤ Ċittadella [S]
⑥ Ġgantija [GD, S, Z]
⑦ Għar Abdul [GD]
⑧ Għar Dalam [GD, S, Z]
⑨ Għar ta' Għeju [GD]
⑩ Għarb [GD, S], x2
⑪ Ħal-Saflieni [Z]
⑫ Il-Haġra l-Wieqfa [GD]
⑬ Il-Latnija [GD?]
⑭ Ix-Xaqqufija [GD]
⑮ Kordin III [GD, S, Z]
⑯ Marfa [GD, Z?]
⑰ Nadur [GD, S?]
⑱ Qala [GD]
⑲ Ras il-Pellegrin [Z]
⑳ San Pawl Milqi [Z]
㉑ Santa Verna [GD, S, Z]
㉒ Skorba [GD, S, Z]
㉓ Ta' Bisqra [GD]
㉔ Ta' Għammar [GD, S, Z]
㉕ Ta' Ħaġrat [GD, S, Z]
㉖ Ta' Kuljat [GD, S, Z]
㉗ Ta' Trapna [Z]
㉘ Tac-Ċawla [GD, S, Z]
㉙ Tas-Sruġ [GD, S, Z]
㉚ Wied ix-Xagħra [S]
㉛ Xagħra Circle [Z]
㉜ Xemxija Hill [Z]
㉝ Xewkija [GD, S, Z]
㉞ Xrobb l-Għaġin [Z]

Figure 3. Map depicting archaeological sites across the Maltese islands where pottery sherds from the Għar Dalam (GD), Skorba (S), and Żebbuġ (Z) phases have been documented in existing literature (Richard-Trémeau *et al* 2023a: 6)

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2.1.2 Skorba and Santa Verna

The sherds from the Għar Dalam phase bearing white-infilled decorations that will be subject to analysis in this research project, were excavated by the FRAGSUS project from Skorba in 2016 (**Figure 4**) and Santa Verna in 2015 (**Figure 5**) (Brogan *et al* 2020, McLaughlin *et al* 2020b, Richard-Trémeau *et al* 2023b, Richard-Trémeau *et al* 2023c).

Skorba

The archaeological site of Skorba is in the north-western Malta, near the village of Żebbiegħ (**Figures 2.22, 4**)
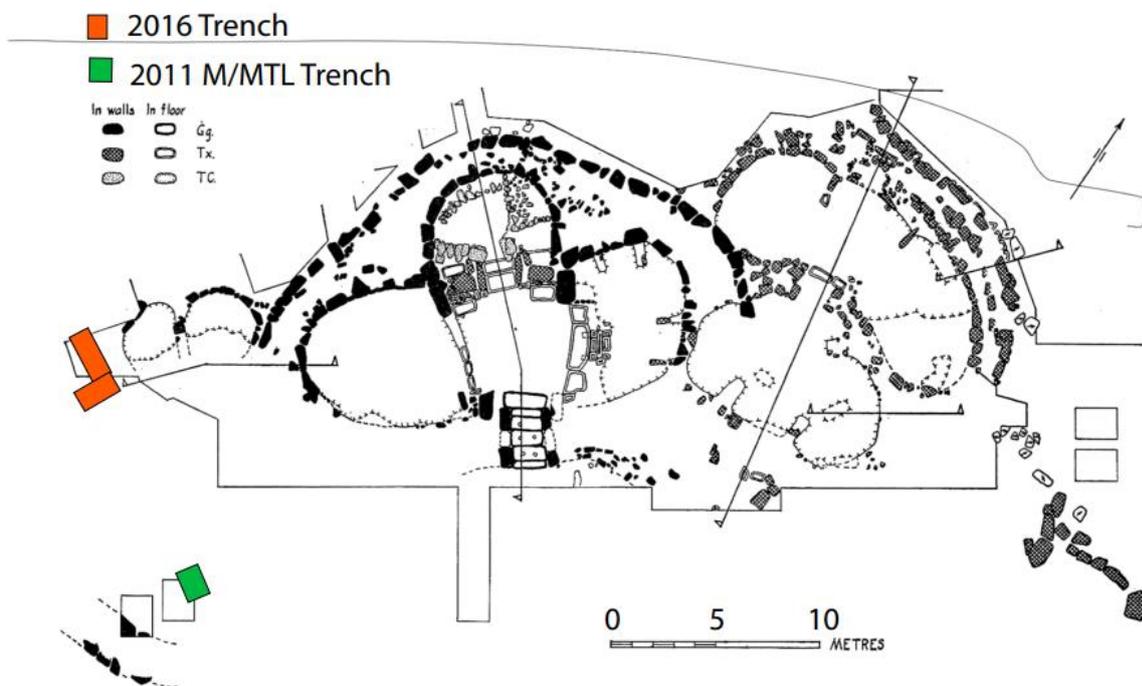
Figure 4: Plan of Skorba as recorded by Trump in 1966, updated to include trenches excavated in 2011 and 2016 (Brogan *et al* 2020: 229)

Positioned at an elevation of 114 meters above sea level, it overlooks L-Imselliet valley and the Binġemma Ridge, and it is near several significant Temple period sites, including Ta Ħaġrat, Xemxija tombs, Tal-Qadi and Buġibba temples (Agius 2022: 27). The presence of fertile land in the northern and southern valleys surrounding the area, along with a freshwater spring in the north and convenient proximity to the coastline, contributed to the site's favourable conditions (Debono Spiteri and Craig 2015: 17).

The site comprises two main structures referred to as the North and South rooms, along with a collection of megalithic constructions (Pirani 2018: 20). Between 1961 and 1963, Trump supervised excavations at Skorba, uncovering a remarkably well-preserved stratigraphic sequence that includes materials from the Early Neolithic through the Bronze Age (Trump 1966: 2). Trump's research introduced pioneering carbon dating methods to the island, leading to the recognition of a new pottery sequence in Maltese prehistory, known as the Skorba phase, bridging the gap between the Għar Dalam and Żebbuġ phases (Agius 2022: 27). Additionally, Trump discovered an oval-shaped hut and a wall constructed from irregular, sizable blocks dating back to the Għar Dalam phase (1966: 10-11).

Skorba underwent additional excavations in 2011, when the Malta Temple Landscapes Project re-excavated and expanded one of the trenches originally opened in 1961 (Brogan *et al* 2020: 230). Further work was conducted in 2016 by the FRAGSUS project, which aimed to collect soil samples from the Early Neolithic layers, providing an in-depth exploration of the site's chronological and environmental context during its earlier periods (Brogan *et al* 2020: 230). One of the aims was to reassess the chronological sequence of the Maltese Islands. Both Trump's excavations and the subsequent FRAGSUS excavations uncovered a stratigraphic record encompassing the entire spectrum of the Neolithic and Temple periods, including layers corresponding to Għar Dalam, Skorba, Żebbuġ, Mġarr and Ġgantija phases (Trump 1966, Brogan *et al* 2020: 229, 240). The FRAGSUS excavation strategically focused on an area anticipated to reveal the Ghar Dalam phase wall identified by Trump (Brogan *et al* 2020: 228). Their efforts revealed Ghar Dalam layers consistently mixed with later sherds (Brogan *et al* 2020: 232-233).

Santa Verna

The megalithic structure known as Santa Verna is located in Xagħra, Gozo. It is found on a high ground close to the edge of an Upper Coralline Limestone plateau, providing extensive views over a large area of the island (McLaughlin *et al* 2020b: 124) (**Figures 2.21, 5**). This site is situated on church-owned land that remains untouched and easily reachable (Malone *et al* 2020c: 14). It currently comprises multiple megalithic blocks forming interior spaces, enclosed by an extensive megalithic perimeter wall (French *et al* 2018) (**Figure 6**).
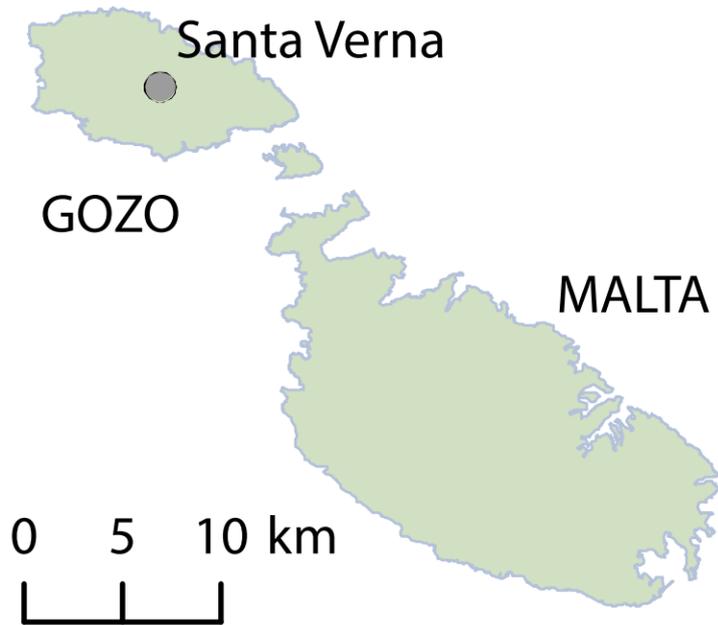
Figure 5: Map showing the location of Santa Verna (McLaughlin *et al* 2020b: 124).
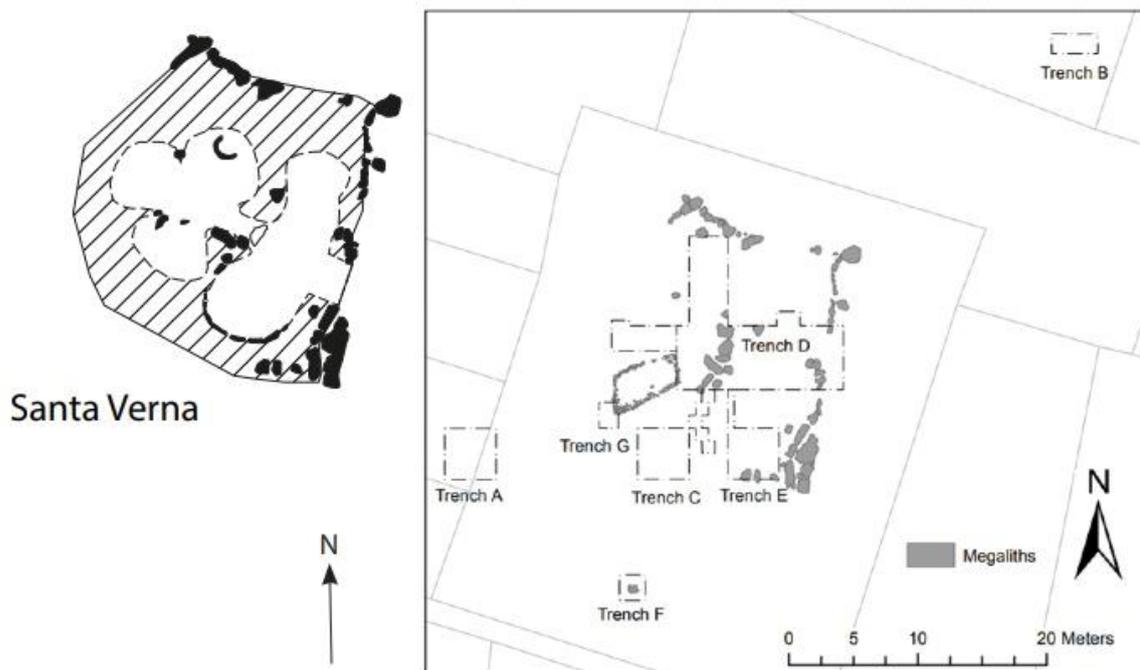
Figure 6: Composite image showing the outline plan of the megalithic structure of Santa Verna (left, adapted from McLaughlin *et al* 2020b: 161) alongside the 2015 excavation trench layout with highlighted major megaliths (right, adapted from McLaughlin *et al* 2020b: 133).

In the early 20th century, the site originally called Torri di Santa Verna was rediscovered by Mr. Nicola Said of the Gozo Public Works Department, who determined its location from an old document (Ashby *et al* 1913: 105). The site was first excavated by the British School at Rome in 1911 (Ashby *et al* 1913: 105-16), and later by David Trump in 1961 (Trump 1966: 19). Trump believed it to be a small trefoil temple, potentially a precursor to Ġgantija, within the remains of an Early Neolithic village (Trump 2013: 195).

In 2014, the FRAGSUS Project surveyed Santa Verna using Ground-Penetrating Radar (GPR) and fieldwalking to identify artifact concentrations in the area (McLaughlin *et al* 2020b: 129). The survey uncovered various prehistoric pottery types close to the site. Għar Dalam pottery was prominent to the northeast and northwest, while Skorba ware was widespread. Various pottery varieties such as Żebbuġ, Mġarr, Ġgantija, Tarxien and Tarxien Cemetery were found in abundance to the temple's west, while Borġ in-Nadur fragments were identified to the northeast. The 2015 excavations at Santa Verna marked the third official fieldwork campaign at the site. Ten trenches, covering a combined area of 160 square meters, were dug at Santa Verna (McLaughlin *et al* 2020b: 123). Most of the unearthed material consisted of topsoil, modern stone rubble assemblages and backfill from previous excavations. The excavation trenches from previous digs were revisited to sample intact archaeological layers with modern methods. McLaughlin *et al* (2020b: 123) confirmed that Santa Verna began as a Żebbuġ phase temple around 3800 cal. BC and it underwent two modifications during this phase, expanding the site into a five-apsed megalithic structure in the Ġgantija phase around 3300 cal. BC. Below the temple, there was a stratum from the Skorba/Għar Dalam phase, suggesting the presence of earlier Neolithic activity. Their excavation revealed the remains of a structure from the Skorba phase, along with various artifacts such as an obsidian arrowhead, clay figurines, chert and obsidian fragments, as well as approximately 23,000 pottery shards spanning different periods in Maltese history, including the prehistoric, Roman and medieval eras. Additionally, radiocarbon dating indicated that fragmented human remains discovered date to the 13th century AD (McLaughlin *et al* 2020b: 123).

2.1.3 Scientific Studies of Għar Dalam pottery

Early scientific studies of Għar Dalam phase pottery were primarily centred on its classification (typology and decoration), with numerous attempts of this conducted over the years, as summarised in **Table 2**. This section will explore the various studies carried outon Għar Dalam phase pottery and the current state of knowledge on the classification, manufacture and provenance of the pottery of this phase. Although this study focuses on the white-infilled decoration, it is essential to explore both the ceramic matrices and the white-infilled decorations of Għar Dalam phase pottery. Examining these elements together is crucial for achieving a comprehensive understanding of the *chaîne opératoire* involved in the production of the pottery.

The initial comprehensive study of Maltese pottery by Evans (1954: 45) briefly mentioned Għar Dalam fabrics and described the sherds as fine wares at that time. Evans (1954: 45-46) distinguished between two types of pottery wares. The first type exhibited a grey hue which occasionally takes on a reddish-brown tint (Evans 1954: 45). This ware has a relatively soft texture and contains a significant quantity of white grits (Evans 1954: 45). The second type was characterised by its compact and well-fired nature, featuring a highly burnished dark-grey or black slip (Evans 1954: 46). Subsequent classifications adopted Evans' categorisation, with Trump (1966) consolidating the two fine wares into a unified category in his study of Għar Dalam pottery found at Skorba. Trump, benefiting from a more comprehensive and well-stratified pottery assemblage at Skorba, also identified coarse wares (Trump 1966: 23). In his classification, fine ware had a matrix in black, grey or brown with a uniform texture and sporadic small gritty inclusions (Trump 1966: 21). The coarse ware exhibited sections that ranged from dark grey to black, whereas the texture was significantly grittier and more prone to crumbling (Trump 2015: 47). Sagona (2015) also adhered to the fabric standards defined by Trump regarding the classification of Għar Dalam pottery.

Ware Type	Evans (1954)	Trump (1966, 2015)	Evans (1971)	Sagona (2015)	Malone et al. (2020a)
Fine	Grey or black	Black, grey, or brown with occasional small gritty inclusions	Soft paste, well-levigated, occasionally has white grits; grey or yellow	Dark to pale grey or brown, refined paste but with some gritty particles	Homogenous with occasional white grits, dark to pale grey or yellow. Thinner walls (average: 8 mm)
Fine	Grey or reddish-brown with fine white grits				
Coarse		Dark grey to black, gritty, friable	Thick and gritty, dark core with a lighter surface, brown, grey, or buff	Dark gritty	Thick and gritty, white grits, crumbles. Thicker walls (average: 12 mm)
Transitional (similar to Skorba fabric)		Gritty fabric			

Table 2: Overview of Għar Dalam pottery categories as described in literature inclusions (Richard-Trémeau et al 2023a: 9)

Trump (1966: 24) also documented a fabric that represented a transitional stage between the Għar Dalam and Skorba phases. In his excavations at Skorba, Trump observed a variant of Għar Dalam pottery, characterised by a fabric similar to the later Skorba pottery, distinguished by its white angular grit (Trump 1966: 24). Such a fabric was discovered in pottery fragments displaying typical Għar Dalam traits, whether in terms of shape or decorative elements (Trump 1996: 24). This variant was identified at Santa Verna (McLaughlin *et al* 2020b: 153), although it was noted that distinguishing it in nondescript fragments lacking distinctive form or decoration may prove challenging, as it closely resembles Skorba fabrics (Richard-Trémeau *et al* 2023a: 8). Furthermore, due to recent excavations by FRAGSUS at this site, which uncovered extensive mixing of Għar Dalam pottery with Skorba phase fragments (McLaughlin *et al* 2020b: 153), the chronological context of this ware cannot currently be confirmed (Richard-Trémeau *et al* 2023a: 8). These examples are relevant as the samples used in the present research project were sourced from Skorba and Santa Verna. Expanding upon these categorisations, Malone *et al* (2020a: 330) noted the difficulty in discerning between pottery associated with the Għar Dalam and Skorba phases. Malone *et al* (2020a) primarily aimed to standardise descriptions of prehistoric Maltese vessels, focusing on aspects such as their form, decorations, style and their distribution across various sites. They maintained the distinction between fine wares and coarse wares in accordance with prior classifications (Malone *et al* 2020a: 330-331).

Molitor's (1988) pioneering research attempted to provide insights into the production techniques employed for the Għar Dalam pottery vessels. Based on petrographic studies, Molitor (1988: 204) contended that the Għar Dalam pottery underwent firing at relatively low temperatures, not exceeding 980°C, in a reducing atmosphere. Additionally, she argued that crushed gypsum was used as a tempering agent in their production (Molitor 1988: 228). While Molitor extensively covered the fine and coarse potteries in her work and acknowledged some

changes in firing methods and adornments (1988: 29-30), she did not address any specific variations in Għar Dalam fabrics or decorations. However, Molitor did make distinctions when discussing Għar Dalam pottery discovered in Għajn Abdul, Gozo, describing this variant as featuring coarse grit within a fine matrix (1988: 228), bearing resemblance to the two fine wares documented by Evans (1954). Unfortunately, Molitor did not provide detailed descriptions of the pottery sherds or the archaeological sites from which they were recovered, making it impossible to re-evaluate these variations. Nonetheless, it is worth noting that Molitor's contributions to our understanding of Early Neolithic pottery include her proposal that intentional clay modification through tempering was a part of the production process (Richard-Trémeau *et al* 2023a: 11).

A study by PhD candidate Scarcella (2011) included Stentinello vessels from Capo Alfiere (Calabria) and Perriere Sottano (Sicily), as well as Għar Dalam phase pottery from Skorba. After extensively reviewing Stentinello phase contexts and published assemblages in Calabria, Sicily and Malta, Scarcella conducted a decoration and typological analysis of assemblages from fourteen sites, including Għajn Abdul, Għar Dalam and Skorba in Malta. Subsamples from three sites were analysed petrographically and using XRD and PIXE methods. For the fifteen sherds from Skorba, Scarcella (2011:193) observed a consistent composition with inclusions of calcite grain, micrite and foraminifera, and differentiated two fabric groups based on the abundance (low and high) of these inclusions. This dissertation is difficult to access and there is no accessible publication reporting these findings.

The Maltese Temples Landscape Project (Allen *et al* 2010) conducted a scientific analysis using portable XRF on eight Għar Dalam phase sherds, along with six sherds from Grey Skorba and ten Red Skorba fragments, all recovered from the site of Skorba. The objective was to determine any differences in the chemical compositions of the ceramic matrix (Allen *et al* 2010: 10). Their results indicated that the three pottery phases, Ghar Dalam, Grey Skorba and Red Skorba, were not easily distinguishable based on the chemical composition of the matrix (Allen *et al* 2010: 10). A possible reason for this could be that the manufacturing techniques employed by the potters close to Skorba may not have undergone significant changes, such as alterations in raw materials, temper or firing methods, resulting in distinct chemical compositions (Allen *et al* 2010: 10). However, some variation in the chemical pattern was noted and it was suggested that this can be understood through the collection of more data (Allen *et al* 2010: 10).

Lastly, Pirone (2017) focused on addressing the origin of some Maltese archaeological artifacts through archaeometry (Richard-Trémeau *et al* 2023a: 11). The utilisation of portable X-ray fluorescence (pXRF) on five pottery fragments from Skorba, all from the Għar Dalam phase, indicated that most of these sherds displayed trace elements similar to those found in clay samples collected from Malta (Pirone 2017: 145). However, there were constraints associated with this research. For example, using a full quantitative approach would have been more suitable for trace element analysis than relying solely on pXRF (Holmqvist 2017: 364). Additionally, Pirone (2017) employed an obsidian calibration material that has been utilised in prior pXRF obsidian research (Tykot *et al* 2013) to analyse comparable trace elements (Th, Rb, Sr, Y, Zr, and Nb). Using a calibration material that is not specifically designed for trace elements in pottery could potentially impact the accuracy and reliability of the study's results. Nevertheless, this preliminary investigation holds importance as it stands as the sole published study that has undertaken a comparative chemical analysis of sherds from various prehistoric Maltese phases and has also identified potential research directions for the examination of local clays. The dispersion observed in Pirone's principal component analysis (PCA) results for the Għar Dalam phase sherds indicated potential chemical disparities among these sherds (Pirone 2017: 146). Pirone (2017: 145) proposed that these variations suggest the utilisation of different clay sources within the Maltese islands.

The increasing number of archaeometric investigations has contributed fresh insights to the conventional examination of pottery, shedding light on various aspects of the pottery record associated with the Maltese Early Neolithic. This progress is made possible by previous archaeological investigations carried out on the sites, as well as advancements in typological and theoretical studies related to material culture. Such foundational work has provided essential resources and potential for further exploration. However, our comprehension of the distinct pottery compositions created by early Maltese potters remains limited (Richard-Trémeau *et al* 2023a: 18). Richard-Trémeau *et al* (2023a: 18, 21) emphasise that while the growing use of archaeometric methodologies and theoretical approaches has enhanced our understanding of Early Neolithic vessel production in Malta, a systematic examination of a series of assemblages using a variety of techniques could help establish localised reference groups. Furthermore, the deployment of these diverse techniques would enhance our grasp of local *chaînes opératoires* and their temporal and spatial variations (Richard-Trémeau *et al* 2023a: 21). These *chaîne opératoires* include all actions carried out by the potters from sourcing the raw material, to decorating and firing the pottery vessels. This is particularly

crucial, as deciphering the different stages involved in the production of Early Neolithic Maltese pottery remains a challenging task based on existing evidence (Richard-Trémeau *et al* 2023a: 21). Finally, Richard-Trémeau *et al* (2023: 21) proposed the undertaking of a nationwide investigation as part of a strategic research plan aimed at advancing our knowledge of archaeological pottery materials, fabrics and *chaînes opératoires*. This initiative seeks to build upon the existing body of knowledge and available materials, while also identifying specific areas for research in typology and fabric analysis, systematically addressing pottery from the Early Neolithic period onward through a series of targeted, "small-scale studies" (Richard-Trémeau *et al* 2023a: 21). One notable research inquiry they highlighted concerns "The study of specific features, such as the decorative white paste on Għar Dalam phase sherds, or the red slip on Skorba phase pottery" (Richard-Trémeau *et al* 2023a: 21). The research proposal put forth by Richard-Trémeau *et al* (2023a: 21) serves as further justification for the present study.

2.1.4 Provenance

The geology of Malta

Chapter 2.2.4 focuses on the different mineral compositions that have been identified in the white infills of impressed pottery from various sites across Europe. This suggests the potential for diverse mineral compositions potentially within the white-infilled decorations of Għar Dalam phase pottery, which is the central focus of this research project. Therefore, it is of importance to conduct a comprehensive review of Malta's geological formations and the minerals they contain, as this information is crucial for interpreting the origin and significance of the compositions found in the white infills of Għar Dalam phase pottery.

The geological structure of Malta consists of a series of sedimentary layers (**Figure 7** and **Table 3**). Beneath these layers, additional strata of sedimentary rocks are present below the sea level, dating back to the late Triassic (Finetti 1985: 215). These consist of dolomite, shales and limestones, which are continuous with North African and South European geological formations (Cilia 2010: 116). These geological features were first identified during a series of oil exploration expeditions conducted in Naxxar and offshore locations, commencing in 1959 (Cilia 2010: 116).
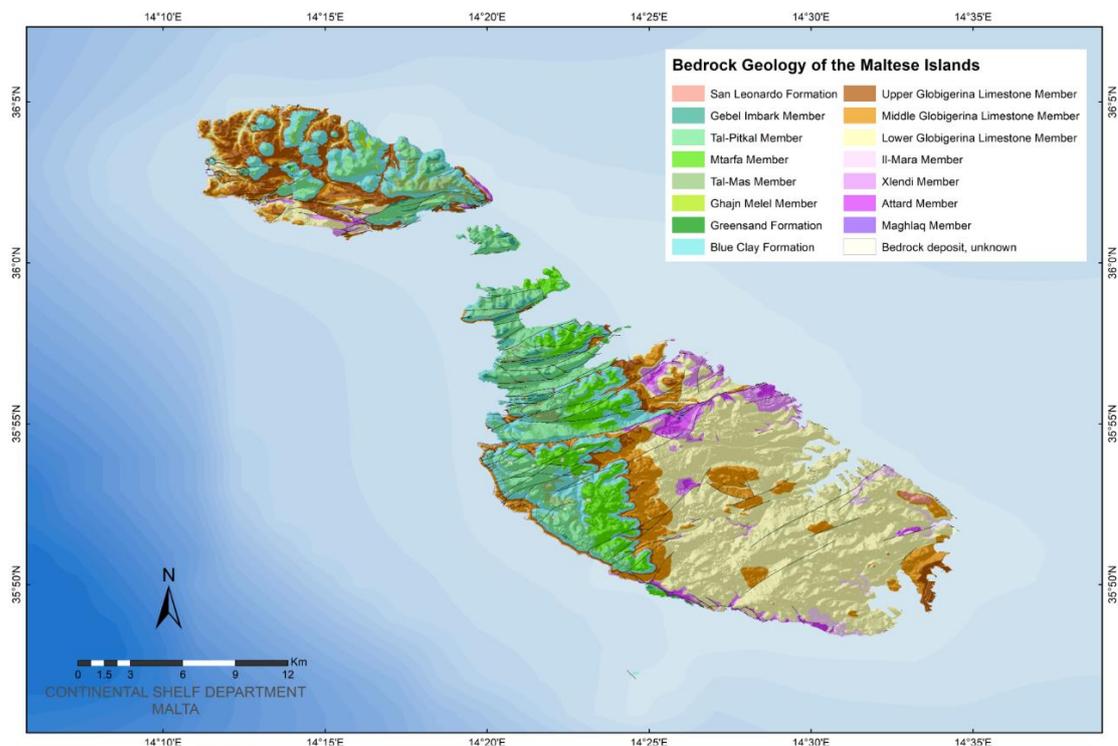
Figure 7: Map of the Geology of Malta (Continental Shelf 2022a)

FORMATION	MEMBER
Upper Coralline Limestone	Ġebel Imbark
	Tal-Pitkal
	Mtarfa
	Għajn Melel
Greensand	
Blue Clay	
Globigerina Limestone	Upper Globigerina
	Middle Globigerina
	Lower Globigerina
Lower Coralline Limestone	Il-Mara
	Xlendi
	Attard
	Magħlaq

Table 3: Geological Sequence of Malta (Continental Shelf 2022b)

The Lower Coralline Limestone, which constitutes the oldest sedimentary layer in the Maltese Islands, is primarily located along the southern coast of Malta, forming imposing cliffs (ERA n.d.). Comprised predominantly of coralline algae, this geological formation suggests its deposition in a shallow gulf setting (ERA n.d.). The Lower Coralline Limestone with an average calcium carbonate ($CaCO_3$) content exceeding 97% (Bianco 2021: 19) is rich in fossil evidence, including shallow-water gastropods with Indo-Pacific affinities (Zammit Maempel 1969: 1). Among the distinctive fossils found are the large *Archaeolithothamnium* coralline algal rhodoliths (ERA n.d.). Additionally, characteristic beds of Scutella echinoids can be found in certain regions, and Terebratula brachiopods are also prevalent in this geological formation (ERA n.d.). These protists' calcium carbonate skeletons contribute to the formation of extensive maerl on the seabed, a substance resistant to decomposition and displacement even after the organisms' demise (Cilia 2010: 116). Over time, these rhodoliths, accumulate and solidify under the influence of overlying pressure and the precipitation of additional calcium carbonate from the surrounding, give rise to expansive deposits of coralline limestone (Cilia 2010: 116). In his study, Cooke (1892) reported the presence of black-coloured Lower Coralline Limestone, possibly containing elevated carbon residues, in the Bengħisa and Għar Ħassan areas.

Globigerina Limestone, the second oldest layer in Malta, is prevalent across roughly 70% of the islands' total land area, contributing to a wide and gently undulating landscape (ERA n.d.). This geological structure is defined by yellow to pale-grey limestones, predominantly comprising the tests of planktonic globigerinid foraminifera (Pedley *et al* 1976: 331). It is divided into Lower, Middle and Upper Globigerina Limestone members, identified by two layers of phosphorite pebbles (Pedley and Clarke 2002: 45). Analysis of Globigerina limestone samples conducted by Murray (1890: 462) revealed a calcium carbonate content ranging from 63.20% to 94.73%. In most instances, magnesium carbonate ($MgCO_3$) was also present, and all samples contained traces of calcium phosphate ($Ca_3(PO_4)_2$) (Murray 1890: 462). Furthermore, Globigerina Limestone may occasionally contain siliceous concretions. These can consist of pure flint, known as silex, while the smaller nodules are composed of chert, a less pure calcareous variety of flint (Groucutt 2022: 6). These concretions can exhibit various colours, including grey or black (Vella 2009: 89). Additionally, limonite is frequently found in areas of Globigerina Limestone formations with a high iron content (Bianco 2021: 13).

Positioned above the Globigerina Limestone, the Blue Clay stratum exhibits a notable tendency to erode when wet, forming talus deposits that cascade over the underlying rock (ERA n.d.). The deposition of Blue Clay may have occurred in a muddy water environment with water depths reaching up to 150 meters, particularly in the lower part of the Formation. Its primary constituents comprise fine lime grains and kaolinite, a clay mineral, giving the layer a compact character when dry and rendering it plastic when wetted. The clay's plastic nature makes it prone to instability and considerable variations in thickness of blue clay deposits have been observed, ranging from 75 meters at Xaghra, Gozo, to a complete absence in eastern Malta. Throughout the Blue Clay stratum, foraminifera are abundant, with species of *Globigerina* and *Orbulina* being the most prevalent. This stratum also contains the remains of marine vertebrates, including *Phoca*, dugongs and various fish. Macrofossils extracted from the Blue Clay strata frequently exhibit impregnation with iron oxide minerals, imparting a brown or orange colour (Cilia 2010: 127). Often present in the Blue Clay are gypsum or selenite crystals, known for their colourless appearance (Bianco 1995: 114). Concretions of goethite $FeO(OH)$, an iron-bearing hydroxide mineral, are present in the Blue Clay stratum (Era n.d.). Furthermore, magnetite (Fe_3O_4), an iron mineral known for its magnetic properties, has also been documented within this layer (Cilia 2010: 148).

The Greensand stratum, found only in select areas of the Maltese islands, is the least common rock formation among the archipelago (Cilia 2010: 127). It consists of bioclastic limestones rich in glauconite, which had been deposited in a warm sea (Pedley *et al* 1976: 334, ERA n.d.). Unweathered sections of Greensand exhibit a green hue but turn to an orange colour upon exposure (ERA n.d.). This deposit is notably thinner than the other four formations, with a maximum thickness of 11 meters observed in localised depressions at Il-Gelmus in Gozo (Pedley *et al* 1976: 334) and in most areas where it is present, it measures less than 1 meter in thickness (ERA n.d.). The indigenous faunal remains found within the Greensand layer are relatively sparse but include *Clypeaster* echinoids, bivalves and local *Heterostegina* (Pedley *et al* 1976: 334). Additionally, remains of Miocene marine fauna are common in Greensand but exhibit phosphatisation or glauconitisation (ERA n.d.). Furthermore, Greensand has been found to contain grains of "silica, lime, clay, and oxides of iron" (Bianco 1995: 115).

The Upper Coralline Limestone, the most recent Tertiary Formation in the Maltese Islands, attains a thickness of approximately 160 meters in the Bingemma area of Malta (Pedley *et al* 1976: 334). In terms of its chemical composition and paleontological characteristics, it

closely resembles the Lower Coralline Limestone, indicating its deposition in shallow marine environments (ERA n.d.). The transition from the Greensand stratum to the Upper Coralline limestone layer above it is gradual, involving sections that merge into "red and black granular sandstone" or "red and white coralline-rich limestone," ultimately transitioning into white calcareous sandstone (ERA n.d.). This sandstone can vary in texture, appearing as compact, soft or porous, but it consistently contains abundant organic remains (ERA n.d.). While certain layers within the Upper Coralline Limestone have become entirely crystalline and have lost traces of their original organic components, other portions are notably fossil-rich, containing well-preserved casts of shells and various other organisms (Pedley *et al* 1976: 334-335).

The five geological formations of Malta are occasionally covered by terrestrial, aeolian and alluvial deposits, which accumulated after the Maltese Islands emerged above sea level during the Quaternary period (ERA n.d.). Quaternary studies have identified a range of such sediments, including tufas, slope deposits, fluvial deposits, palaeosoils, coastal deposits, aeolian silts, as well as cave, fissure, and doline deposits (Hunt 1997: 101).

Scientific Studies of Maltese Blue Clay

In addition to classification, discussed in **Chapter 2.1.3**, the analysis of pottery fabric compositions can offer valuable insights into the origins of the constituents utilised in the production of pottery vessels (Richard-Trémeau *et al* 2023a: 4). To identify the origins of clay samples in a regional investigation, it is presupposed that the differences among various clay sources are more significant than the variations within a single source of clay (Richard-Trémeau *et al* 2023a: 4). This assumption gains support from a number of studies (Visser 1992, Montana *et al* 2011: 487), which showed that the presence of clay sources in certain areas of Sicily could be discerned based on the varied geological substrates underlying the region. On the other hand, within the Maltese archaeological context, researchers have encountered challenges stemming from the relatively uniform geological composition of the region (Richard-Trémeau *et al* 2023a: 4). For instance, Molitor (1988: 160) employing XRD analysis failed to unveil significant disparities between clay sources in Malta and Gozo. The full extent of chemical and mineral differences in the blue clays has yet to be thoroughly investigated. Nonetheless, Pirone (2017) emphasised that chemical differences within a single clay source, as observed through the analysis of clay samples from various elevations on the slopes in both Selmun and Ġnejna, may be more pronounced than those found between different clay sources.

The hypothesis positing potential compositional similarities in clay elemental composition between Malta and Ragusa (Sicily), as suggested by Bruno (2009: 109), remains untested. Notably, DiGeronimo *et al* (1981) had elucidated resemblances in geological formations between Malta and the southeastern part of Sicily. In a broader context, research endeavours should address the chemical and mineral differences in indigenous sediments while evaluating their adequacy for pottery production (Richard-Trémeau *et al* 2023a: 5). Until such investigations are undertaken, the composition of vessel fabrics can only be assessed for consistency with Maltese origins through comparative analyses of various Maltese clays and temper sources with archaeological pottery (Richard-Trémeau *et al* 2023a: 5).

2.2: White-infilled Decorations

2.2.1 Why Decorate Pottery?

Understanding the motivations behind pottery decoration necessitates engagement with anthropological theories and ethnographic research that elucidate the cultural and social dimensions underpinning these practices. As such, this section will focus on different studies describing why people decorate pottery, exploring various theoretical perspectives and findings that highlight the significance of decorative traditions.

Boas (1955) argued that potters readily decorate pottery whenever the opportunity arises, exploiting the malleable nature of clay (Braun 1991: 365). Furthermore, the intricate process of pottery making, marked by various steps demanding time and skill (*chaîne opératoire*), inherently lends itself to diversifying decorative elements (Braun 1991: 365). Since the additional incorporation of decoration, unless it involves intricate designs, may not substantially extend the temporal and labour investment, potters can be more inclined to adopt such embellishments (Braun 1991: 365). Nevertheless, potters might refrain from decorating vessels utilised for mechanical purposes, owing to the functional context they anticipate (Braun 1991: 365).

Certain theories in archaeology, including interaction theory and information exchange theory, converge on the notion that decoration, along with other aspects of artefact form, can be used as cost-effective conduits for conveying messages (David *et al* 1988: 365). The varying aspects encompass the content of these messages, the sender and recipient identities, the degree of intentionality and the interpretive lens of the audience (David *et al* 1988: 365). As an example, David *et al* (1988) discuss the utilisation of pottery among the Mafa and Bulahay communities to convey collective messages. They explain that these decorations affirm social order and serve as mnemonic visual expressions of cultural beliefs and thoughts, highlighting their role beyond mere functionality. Moreover, the authors hypothesise that people extend certain concepts and treatments accorded to individuals to pottery, recognising fundamental similarities in transformation and other resemblances. They contend that the human body, being an essential medium of expression, influences much of the decorative expression found in pottery (David *et al* 1988: 366). The association of pot to person according to David *et al* (1988), makes pottery decoration a valuable tool for ethnographic and archaeological studies. They argue that the decoration on pottery not only embodies explicit messages but also carries implicit components that reflect societal values and religious beliefs (David *et al* 1988: 379).

Contrary to the notion of pottery decoration as mere ornamentation or emblematic of ethnicity, the authors assert that it serves as a "low-technology channel" through which societies imbue their values in people (David *et al* 1988: 379). For example, among the Mafa and Bulahay, pottery is decorated with motifs that symbolise protection against fate and dangers associated with authority. These motifs, deeply embedded with cosmological and religious significance, demonstrate how pottery decoration in these cultures transcends aesthetics to reflect and reinforce societal values and beliefs, serving as both protection against existential threats and insulation against the inherent perils of authority, whether originating externally or from within the vessel (David *et al* 1988: 365).

In a commentary in the same paper by David *et al*, Whitney Davis challenges the assertion that decorations "can be cost-effective means of sending messages" (David *et al* 1988: 365), deeming it "partial and somewhat misleading" (Davis 1988: 380). Davis (1988) contends that style can possess an iconographical or denotational aspect. Additionally, she argues that David *et al* (1988) offer a perspective influenced by functionalist anthropology. She highlights the complexity of empirically analysing the process of communication through symbolic morphology. This recognition underscores the challenge in ascribing clear intentions and meanings to symbolic forms. Moreover, Davis draws attention to the limitations of viewing communication as the sole or primary function of symbolic morphology. Drawing from Chomsky's critique (Chomsky 1975: 56-77) of Grice's theory of linguistic meaning (1957), which emphasises the social intention to produce a response in the audience, Davis suggests that communication is just one aspect of symbolic expression. Furthermore, Davis contends that part of style may lack a specific audience, indicating that not all symbolic forms are created with the explicit purpose of communication. Moreover, she challenges the notion that all cultural pathways lead to social communication, suggesting that the history of production, including the creation of symbolic forms, is not entirely a social history. This perspective implies that attributing solely communicative functions to symbolic morphology may oversimplify its role within cultures. Davis (1988: 380) also raises questions on the source of the message portrayed. While David *et al* (1988) suggest that pottery decoration may be influenced by conceptions of the body, they caution against regarding this as the sole explanation. They suggest that pottery decoration, alongside other cultural practices like body ornamentation, ironworking and architecture, may originate from a cultural pattern, albeit one that remains elusive. Davis expresses unease with the functionalist-structuralist approach employed by David *et al* (1988), highlighting its tendency to overlook inquiries into historical

genesis and evolution. It contends that the history of symbol generation and replication does not necessarily require specific origins or a predetermined position within an existing framework of forms: "although all "structures" of meaning have history, not all history of meaning has "structure"" (Davis 1988: 380).

The perpetuation of decorative traditions hinges upon the transmission of knowledge and skills from one generation to the next (Braun 1991: 365). Cultural shifts impacting this intergenerational passage can influence the richness of decorative practices (Braun 1991: 365). Additionally, the dichotomy between pervasive and partitive decorative behaviour offers insights into the extent and coherence of ideological themes (DeBoer 1991), which could also affect embellishments. Pervasive decoration represents ideological elements central to a community's identity, whereas partitive decoration lacks such ideological cohesion (DeBoer 1991: 148-151). Consequently, changes in either type of decoration prompt questions about their underlying causes (Braun 1991: 365). On the other hand, it has been also proposed that artisans are "active users and often innovators rather than simply passive receivers of their knowledge and skills" (Braun 1991: 366). In social contexts marked by uncertain interpersonal dynamics, the act of decoration serves as a tool for navigating complex interactions (Braun 1991: 366). When social uncertainty is elevated (e.g. political or economic instability), there is an anticipation of an increased emphasis on decoration, as individuals endeavour to shape perceptions through visual means (Braun 1991: 367). Conversely, in environments where opportunities for visual communication are limited due to factors such as reduced interaction, limited physical visibility or the lack of social tensions, one would expect to observe less decoration (Braun 1991: 367). The suitability of utilitarian objects for communication within their specific usage context plays a pivotal role in this dynamic. Consequently, alterations in settings in which these objects are employed could potentially influence the degree of decoration (Braun 1991: 367). Moreover, the visibility of various objects to different social groups necessitates a comprehensive exploration of the social dynamics that emerge within diverse physical contexts (Braun 1991: 367).

Tsetlin (2006) employs a "historical-and-cultural scientific approach" to investigate the formation processes of each component within the creation of decorations, which consists of three main aspects: technical and technological, stylistic and semantic (Tsetlin 2006: 1). The technical and technological component explores the origins of various methods of pottery decoration, the stylistic component delves into the development of decorations, and the

semantic component focuses on understanding the underlying meanings of the decorations. Tsetlin (2006: 7) outlines the evolution of pottery decoration as a multifaceted process, characterised by two primary approaches: firstly, the development of pottery vessels with "technological-and-decorated surfaces," which served dual functions of functionality and aesthetics, enabling users to differentiate their vessels from others; and secondly, the intentional embellishment of clay vessels, reflecting a more advanced stage in pottery decoration, where new designs were created through deliberate surface adornment. Finally, Tsetlin (2006: 7) suggests that incised or carved decoration was primarily linked with durable materials such as stone, bone and wood. This suggests that incision as a decorative practice likely predates pottery production and subsequently influenced pottery-making, possibly through the transmission of techniques by craftspeople accustomed to working with non-clay materials like stone vessels.

The research reviewed highlights the diverse purposes and interpretations surrounding pottery decoration, illustrating its significance in both artistic expression and cultural communication. Pottery decoration serves as a multifaceted phenomenon encompassing both functional and symbolic dimensions across various cultural contexts. Braun (1991) argues that embellishing pottery enhances its aesthetic appeal without significantly increasing production effort, leveraging the malleability of clay to explore diverse decorative avenues. Similarly, David *et al* (1988) assert from an ethnographic and archaeological perspective that pottery decoration functions as a medium for transmitting cultural messages and reflecting societal values, elucidating how decorative motifs encode explicit messages and implicit cultural meanings. Davis (1988) critiques functionalist perspectives by challenging the notion of decoration solely as a cost-effective means of communication, emphasising the complexities of interpreting symbolic forms. Additionally, Tsetlin (2006) provides a historical perspective, tracing the evolution of pottery decoration through technical, stylistic and semantic lenses, highlighting its dual roles in functional differentiation and intentional aesthetic expression.

2.2.2 Cardial and Impressed wares

Cardium pottery, also known as Cardial ware, represents a distinct Neolithic decorative style characterised by clay being imprinted with the shells of *Cardium edule*, a member of the *Cardiidae* cockle family (**Figure 9**) (Sagona 2015: 23). This type of pottery serves as a defining feature of the Neolithic style responsible for its creation and diffusion, commonly referred to as the Cardial culture. Alternatively, some archaeologists employ the term 'impressed ware' to describe this cultural phenomenon, as the impressions on the pottery can be made using sharp objects rather than cockle shells, such as nails or combs (Oxford Reference 2023). Impressed ware (**Figure 10**) is primarily found in the region stretching from Italy to the Ligurian coast (Capelli *et al* 2017: 532), distinguishing it from the western expansion of Cardial pottery, which extends from Provence to western Portugal (**Figure 8**) (Guilaine 2018: 212).

Figure 8: Map depicting the different distributions of Early Neolithic pottery cultures (6th millennium BC). Areas marked with question marks represent locations where archaeological evidence of the Neolithic is anticipated but remains undiscovered (Keerl 2014: 5).

The earliest instances of impressed ware, dating back to 6400-6200 BC, are situated in Epirus and Corfu (Cunliffe 2008: 115-116). Subsequently, settlements with impressed ware emerged along the eastern Adriatic coast in Albania and Dalmatia, with origins between 6100 and 5900 BC (Cunliffe 2008: 115-116). Additionally, Italy's earliest known occurrence of impressed ware is associated with Coppa Nevigata, located on the southern Adriatic coast, with a potential dating as early as 6000 cal BC (Natali 2014: 2). Furthermore, evidence of early

manifestations of Cardial pottery dating back to around 6000 BC is also apparent during the Su Carroppu culture in Sardinia (Lugliè *et al* 2007: 430). Moving northward and westward, all securely dated radiocarbon evidence aligns with dates found in Iberia around 5500 cal BC, pointing to a swift expansion of Cardial and Impressed wares, spanning approximately 2,000 kilometres from the Gulf of Genoa to the Mondego estuary, likely occurring within a span of no more than 100-200 years, at a rate of 10-20 kilometres per year (Zilhão 2001: 14184). This suggests a maritime-driven expansion, involving the establishment of colonies along the coastline (Zilhão 2001: 14184).
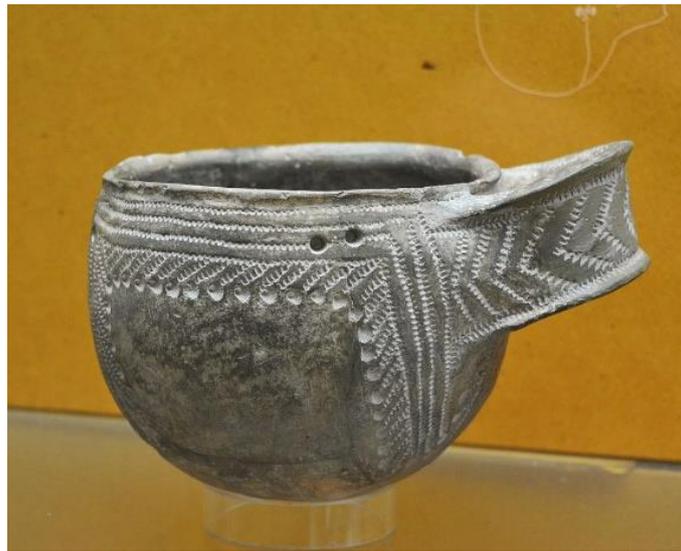
Figure 9: An example of a Cardium Ware pot from Cova de l'Or, Spain (5,000-4,200 BC) (Joanbanjo 2013).

Figure 10: An example of an Impressed Ware pot from central Dalmatia (6,000-5,500 BC) (McClure *et al* 2014: 1021).

2.2.3 White Infill and its Relation to Impressed Ware

Infills were a common decorative element found in incised or impressed pottery throughout Europe, spanning from the Neolithic period to the Iron Age, and encompassing regions from the Iberian Peninsula to the Balkans (Odriozola 2008: 41). These infills exhibited diverse mineral compositions and colours, including red, yellow and white (Pessina and Tiné 2008: 81). This diversity in infills can be attributed to variations in raw materials and production techniques (Odriozola 2008: 41).

A method for applying infill to pottery decorations may have involved the following steps. The production process began with the selection of materials, such as bones (Odriozola *et al* 2012: 149) or calcium carbonate (Roberts *et al* 2008: 322), although other materials were used. These materials were ground to create a paste (Odriozola *et al* 2012: 149). The paste was formed by mixing the ground material with water, and in some cases, animal fats or bone marrow (Odriozola *et al* 2012: 149). This mixture created a substance that could be applied to the incised or impressed decorations. Once the vessel had dried and ready for infilling, the paste was carefully applied into the incisions/impressions (Odriozola *et al* 2012: 149). Subsequently, the vessels were fired (Odriozola et al. 2012: 149). It is worth noting that the firing process could impact the materials used. For instance, bone undergoes physicochemical changes during firing (Odriozola *et al* 2012: 149), and calcium carbonate may dissociate if exposed to high temperatures (850°C) or prolonged firing (Odriozola *et al* 2012: 149). Because of these considerations, it has been suggested that infilling could potentially have been carried out after the firing process, rather than before, to better ensure a successful production outcome (Odriozola *et al* 2012: 149).

2.2.4 Scientific Studies of White-infilled Decorations

The archaeological literature discusses the practice and tradition of filling incisions and impressions on pottery with infill material. While the technique's origin and diffusion have received limited research attention, there is a substantial body of scholarly work dedicated to the archaeometric analysis of these infills. In this section, scientific studies on white-infilled decorations in Neolithic Impressed pottery will be initially reviewed, given their relevance to the Għar Dalam phase pottery. Following this, further scientific studies analysing white-infilled decorations from other regions of Europe within a similar timeframe will be examined. This approach will provide a comprehensive review of scientific studies conducted on white-infilled decorations from various pottery cultures, elucidating the diverse compositions found within these infills, thus providing insights into the composition(s) that can be found in the white-infilled decorations of Għar Dalam phase pottery, and it will serve as a basis for the methodology used in the present study.

The section will commence with a reference to a study conducted on the white-infilled decorations of Stentinello pottery, given its association with Għar Dalam phase pottery. Subsequently, other studies on white-infilled decorations will also be reviewed. Scarcella *et al* (2011) analysed Stentinello pottery with white-infilled decorations from Capo Alfiere in Calabria and Perriere Sottano in Sicily (**Figure 11**).
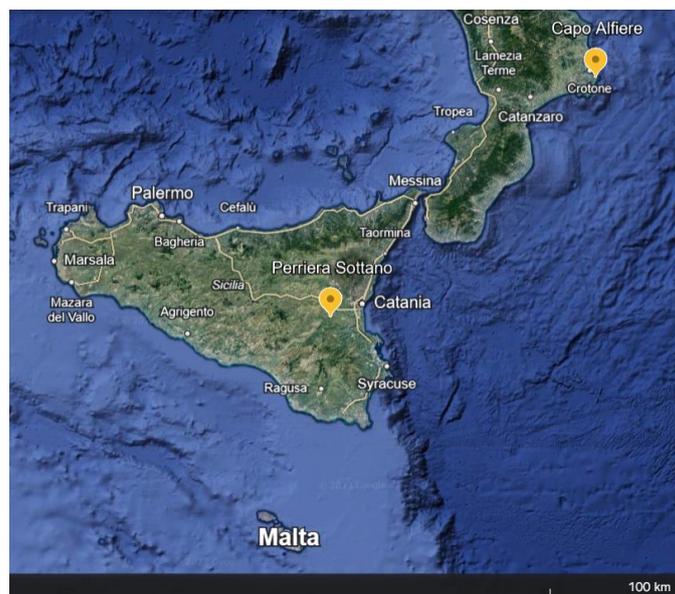
Figure 11: Map showing the geographic locations of Capo Alfiere and Perriere Sottano (Google Earth 2023)

Capo Alfiere, situated at the northeastern edge of Calabria, is an archaeological site known for its production of Stentinello ceramics, which date to the 5th millennium BC (Morter 1992: 28). Perriere Sottano, an archaeological site in the commune of Ramacca, Sicily, has also yielded fragments of pottery featuring impressed decoration, characteristic of the Stentinello style from the 5th millennium BC (Freund *et al* 2015: 208).

Very few scientific studies have been conducted on the white-infilled decorations of Stentinello phase pottery. One study on this matter was conducted by Scarcella *et al* (2011). For their analysis, Scarcella *et al* (2011: 156) selected eight pottery samples from Capo Alfiere and three from Perriere Sottano. Preliminary petrographic examinations revealed that all the infills consisted of "micritic calcitic paste" (Scarcella *et al* 2011: 156). However, differences were observed between the white infills of Perriere Sottano and Capo Alfiere. While calcite was the dominant mineral in both, their structures differed (Scarcella *et al* 2011: 156). The samples from Perriere Sottano exhibited a fine micritic paste, whereas the samples from Capo Alfiere displayed a more crystallised calcium carbonate (Scarcella *et al* 2011: 156) (**Figure 11**). This variation could have caused by disparities in the preparation of the white infill or potentially, because of distinct burial conditions that facilitated secondary crystallisation (Scarcella *et al* 2011: 157).

Additionally, Scarcella *et al* (2011: 157) also discussed variations in the fabrics of the sherds. They observed that the fabrics displayed a varying size distribution of temper and feldspars relative to quartz (**Figure 12**).
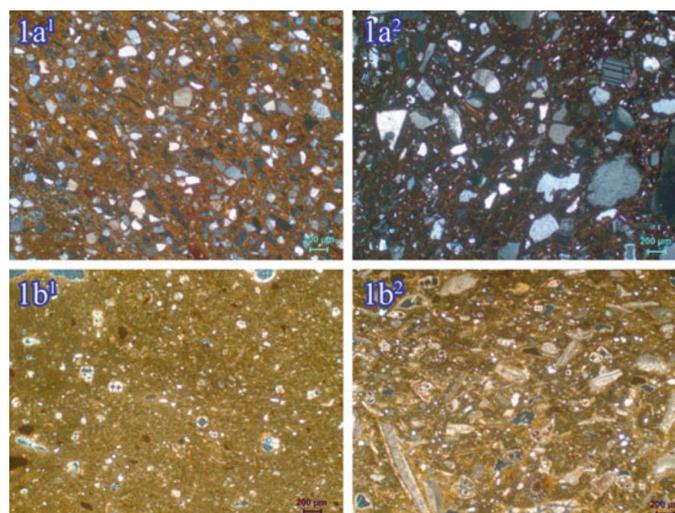
Figure 12: Photos of thin sections of pottery samples from Capo Alfiere (1a[1] and 1a[2]) and Perriere Sottano (1b[1] and 1b[2]), showcasing the size distribution differences between the two (Scarcella *et al* 2011: 155).

Furthermore, the fabrics from Capo Alfiere and Perriere Sottano exhibited distinct mineralogical and chemical compositions, which facilitates their differentiation. The authors attributed these differences to the sourcing of materials, suggesting that each village procured clay from the nearest available clay outcrops (Scarcella *et al* 2011: 157).

The findings presented in the paper by Scarcella *et al* (2011) are derived from her PhD research (Scarcella 2011). Besides the results included in the paper, her PhD research also involved other analyses. In her PhD, Scarcella (2011) examined the infilled decorations of Stentinello pottery from Capo Alfiere and Perriere Sottano, as well as a Għar Dalam phase sherd from Skorba (MAL 7). The infills exhibited different colorations: six samples had white-infilled decorations (CAP 1, CAP 6, PER 1, PER 7, PER 12 and MAL 7), three had red infills (CAP 12, CAP 13 and CAP 23) and the other three had yellow colourations (CAP 9, CAP 18 and CAP 19) (Scarcella 2011: 208). All samples underwent chemical analysis using EDS, but only three (CAP 12, CAP 23 and PER 7) underwent mineralogical analysis using XRD (**Table 4**).

Sample	Colour	SEM	XRD
CAP 1	White	*	
CAP 6	White	*	
CAP 9	Yellow	*	
CAP 12	Red	*	*
CAP 13	Red	*	
CAP 18	Yellow	*	
CAP 19	Yellow	*	
CAP 23	Red	*	*
PER 1	White	*	
PER 7	White	*	*
PER 12	White	*	
MAL 7	White	*	

Table 4: Table adapted to English from Scarcella (2011: 208) showing the sample list, including the colour of the infills and the scientific analyses carried out.

The chemical elements constituting the coloured infills of the ceramics were determined through a series of EDS analyses, while XRD identified the minerals present. This determined that the samples contained a mixture of calcium carbonate and clay (Scarcella 2011: 208-209). The hue of the coloured infills from Capo Alfiere seemed to vary based on the ratios between calcium carbonate and iron minerals: white infills contained a higher percentage of calcium carbonate compared to iron, while the red ones had higher iron content relative to calcium carbonate. Although calcium carbonate was consistently present, its form varied by origin. Fragments from Perriere Sottano and the sample from Skorba were characterised by very fine micrite, whereas the calcium carbonate in fragments from Capo Alfiere is more crystallised.

The XRD analysis produced the following results. Sample CAP12 showed proportions of quartz and calcite, while sample CAP23 was dominated by quartz. The latter sample also contained traces of hematite, explaining the red colour of the infill. Finally, sample PER7 exhibited a similar composition to CAP12. Observations regarding the manufacturing process were made. In all cases, the basis for the production of the coloured pastes was a mixture of calcite and clay. The calcite was ground to varying degrees of fineness (finer in Perriere Sottano than in Capo Alfiere). However, the crystallisation of calcite in Capo Alfiere could have resulted from chemical alteration during burial. At Capo Alfiere, varying percentages of hematite were added to the calcite and clay mixture to achieve red or yellow pastes. Additionally, Scarcella (2011) noted that the application of the pastes varied (**Figure 13**). Some pastes were applied throughout the depth of the impressions or incisions, occasionally extending beyond the recesses and creating a spread effect over the entire decoration. Additionally, in certain cases, the application exceeded the depth of the recesses, suggesting that part of the paste was removed during the production process. Scarcella (2011: 211-212) noted that the application of coloured pastes in the recesses of the impressions sometimes resulted in a deformation of the contours, suggesting that the infills were likely applied before the firing of the vases.
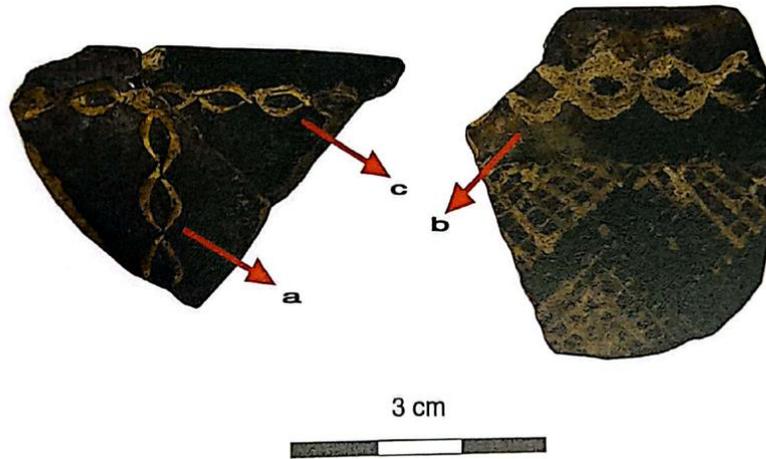
Figure 13: The different types of applications noted by Scarcella (2011). a. within the impressions/incisions, b. spread over the impressions/incisions, c. deformation in the contours of the decorations.

The methodology adopted by Scarcella (2011) and Scarcella *et al* (2011) is well-suited and established for the scientific analysis of coloured infilled decorations. Polarised Light Microscopy (PLM) has been effectively used in pigment examination (Burgio and Clark 2000, Cheng *et al* 2007). Under polarised light, minerals display distinctive optical properties such as colour, birefringence and pleochroism, which aid in their identification. Additionally, SEM-EDS has been a staple in pigment analysis, providing high-resolution imaging and elemental composition analysis (Genestar and Pons 2005, Lluveras *et al* 2010). XRD has also been an established main analytical technique, used to gather mineralogical data for pigment analysis (Parras-Guijarro *et al* 2006, Senvaitiene *et al* 2007, Romano *et al* 2011). The studies by Scarcella (2011) and Scarcella et al. (2011) adopted a multi-analytical approach, which enabled the proper analysis of white-infilled decorations. However, these methods were not all applied to the fragment from Skorba.

Another study on the infilled decorations of Italian impressed pottery was conducted by Giustetto *et al* (2013). This study encompassed periods ranging from the Neolithic to the Bronze Age. Its aim was to determine the constituent materials of decorative pottery infills discovered at the archaeological site of Castello di Annone in Piedmont, North-Western Italy (**Figure 14**) (Giustetto *et al* 2013: 4249).
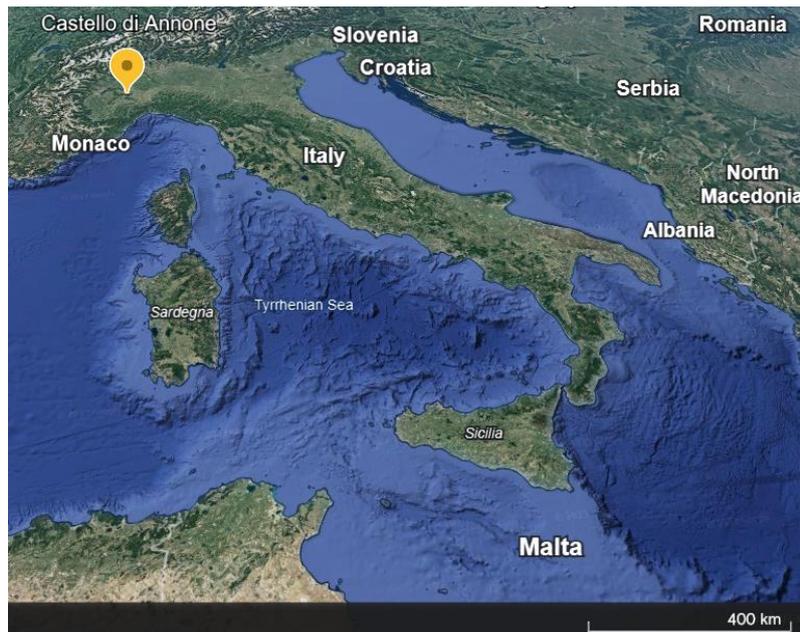
Figure 14: Geographic location of Castello di Annone (Google Earth 2023)

Situated on a hill district alongside the Tanaro River, the Castello di Annone site witnessed multiple occupational phases spanning from the middle Neolithic (4600 BC) to the middle Iron Age (6th-5th century BC) and extending into the post-medieval epoch (Giustetto *et al* 2013: 4250). Twenty-nine Neolithic age impressed pottery sherds adorned with white-infilled decorations, were selected for scientific analyses (Giustetto *et al* 2013: 4250). Of these, 27 belonged to the phase II of the Vasi a Bocca Quadrata culture (4600-4200 BC), which are characterised by a meander-spiralic style epoch (Giustetto *et al* 2013: 4250). This phase can be categorised into two groups: the first comprises the finer vessels, including square or round-mouthed bowls adorned with decorations exhibiting shapes such as excised triangles; the second group encompasses round-mouth shapes (bowls, vases and cinerary urns) characterised by "notched rims and/or wall-dragging decorations" (Giustetto *et al* 2013: 4250). The influx of people from the low and medium Rodano valley, occurring between 4400 and 4200 BC, brought with it Western features that influenced and blended with the traits of the Vasi a Bocca Quadrata culture (Giustetto *et al* 2013: 4250). The remaining two sherds used in this study were from this period. This ceramic style can also be divided into two distinct groups. The first group displays traits resembling the Saint Uze style (4500 - 4000 BC), characterised by deep shapes while the second group exhibits decorations typical of the Chasséen Culture (4500 - 3500 BC), including "painted lattice-disposed triangles, bands filled by vertical lines, seldom with irregular geometric motifs" (Giustetto *et al* 2013: 4250). **Figure 15** displays some Neolithic sherds used in the study.

Figure 15: Pottery fragments adorned with white-infilled decorations from Castello di Annone (Giustetto *et al* 2013: 4251).

A comprehensive archaeometric analysis was conducted utilising a multi-disciplinary, micro-destructive approach that incorporated Fourier-Transform Infrared Spectroscopy (FTIR), Raman spectroscopy and X-Ray Powder Diffraction (XRPD). The recurring presence, in several FTIR spectra, of the δ(OH) absorption band at 669 cm^{-1} leads to the possible presence of talc [$Mg_3Si_4O_{10}(OH)_2$] (Wilkins and Ito 1967: 1658-1659). In the study by Giustetto *et al* (2013: 4256), talc signatures were unequivocally detected in 13 out of the 29 Neolithic sherds analysed through FTIR. Other samples exhibited the presence of a distinct type of hydroxyl group, with absorption bands identified at 3572 ν(OH) and 630 cm^{-1} δ(OH). These were accompanied by bands indicative of the presence of CO_3^{2-}, as evidenced by signals at 1460, 1419 and 873 cm^{-1}, as well as PO_4^{3-} groups observed at 1040, 1093 and 962 cm^{-1} (Giustetto *et al* 2013: 4254). Additionally, a reading at 1640 cm^{-1} was attributed to δ(H$_2$O), while its stretching counterpart manifested as a broad band between 3250 cm^{-1} and 3500 cm^{-1} (Giustetto *et al* 2013: 4254). These findings indicated the potential presence of a phosphate/carbonate phase, exemplified by hydroxyapatite [$Ca_5(PO_4)_3(OH)$] and carbonate-hydroxyapatite [$Ca_{10}(PO_4)_5CO_3(OH)$], which are mineral constituents of bone (Giustetto *et al* 2013: 4254). This implies the plausible utilisation of Bone White, an ancient pigment derived through the calcination of animal bones, teeth and antlers (Giustetto *et al* 2013: 4254). **Figure 16** displays the FTIR spectra of two samples, one predominantly composed of talc and the other of Bone White.
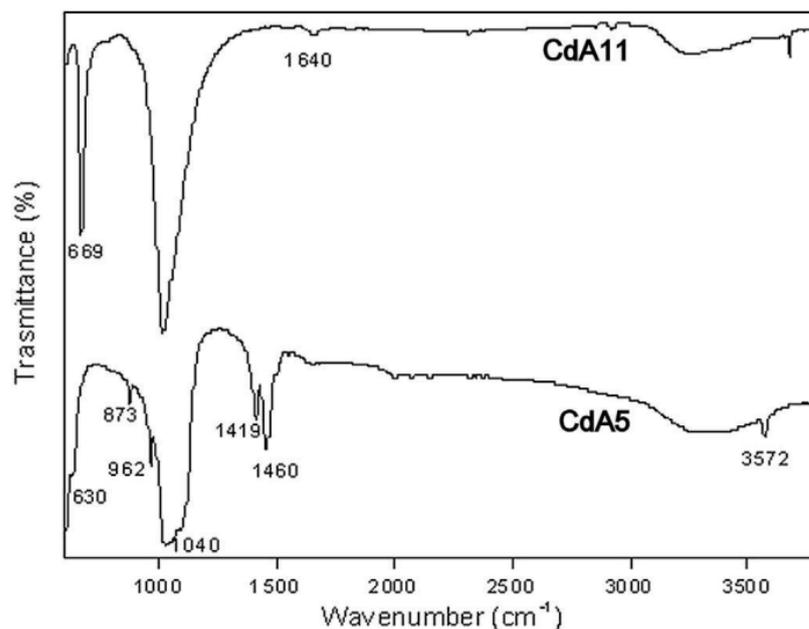
Figure 16: FT-IR spectra of CdA11 (talc) and CdA5 (Bone White) (Giustetto *et al* 2013: 4254).

Strong fluorescence was observed in the Raman spectra of all analysed samples, hindering a straightforward interpretation of the collected data (Giustetto *et al* 2013: 4254). Nonetheless, the presence of Bone White was confirmed by the presence of a phosphate band, consistently emerging at approximately 961 cm^{-1}. This observation was particularly notable in specimens showing a recurrence of the typical hydroxyapatite IR absorption band. In certain samples, FTIR absorption bands associated with both talc and hydroxyapatite were identified, suggesting the potential use of a mixture of these two materials in creating the white infills. The assignment of each signal to a specific colouring agent proves challenging, as bands related to the predominant component tend to overshadow those of the secondary material, manifesting only as weak shoulders. Infrared signals indicating the potential presence of calcite, likely attributed to secondary post-burial deposition processes, were observed on two occasions. In certain instances, spectroscopic data collected from the white infill closely resembled those of the ceramic body, hinting at possible contamination during sampling. Despite this limitation, arising from the limited amount of white infill in the sample, it generally did not hinder the accurate characterisation of the white constituents. Weak bands, likely associated with the presence of proteins, were also observed. While unequivocal identification of the source for these proteins proved challenging, these signals may be attributed to traces of binding agents that facilitated the adhesion of the white infills to the ceramic body. In specific cases, traces of a paraloid, a resin, were also detected, indicating the consequence of a prior restoration intervention.

The increased sensitivity provided by XRPD in detecting bulk composition facilitated a comprehensive characterisation of the minerals comprising the infilled decorations. This analysis not only revealed the presence of all minerals associated with the ceramic body but also confirmed the utilisation of essentially two distinct constituents in the manufacture of the white-infilled decorations (Giustetto *et al* 2013: 4254). Based on their mineralogical composition, the white-infilled decorations were broadly categorised into three main groups: a) talc-constituted infills (9 samples); b) those with a combination of talc and hydroxy-/carbonate-hydroxyapatite (bone ash) (13 samples), either as the sole component (12 samples) or in combination with an additional constituent (1 case, CdA27, had kaolinite); and c) Bone White (3 samples). An exception to this classification was CdA27, which exhibited calcite + kaolinite make-up. Unequivocal characterisation was not achievable for four samples due to either insufficient sampling or significant contamination from the ceramic body during sampling. XRPD patterns for these categorisations can be observed in **Figure 17**.
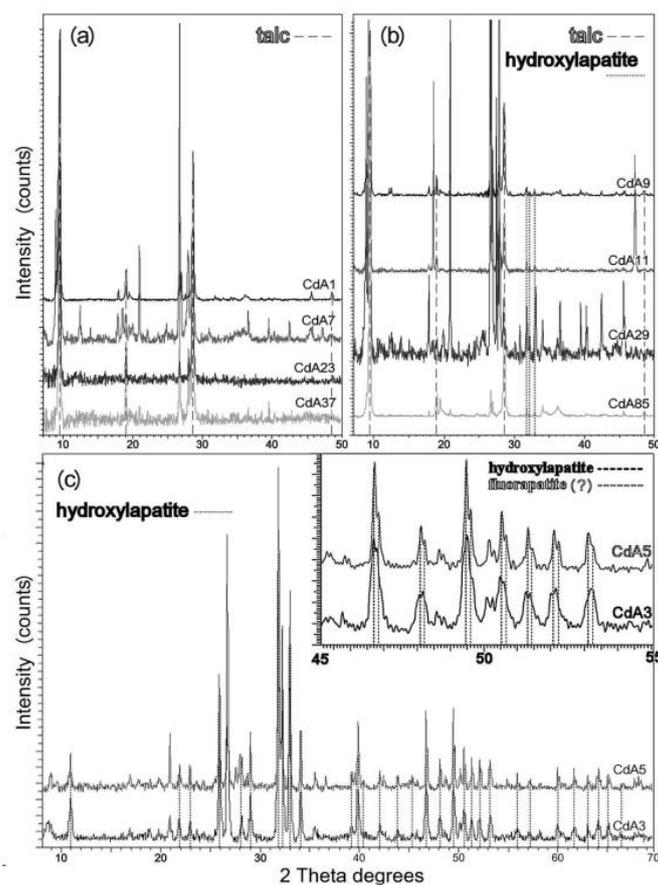
Figure 17: XRPD patterns of white-infilled decorations in Neolithic pottery from Castello di Annone. a. Talc-based white-infilled samples. b. Combination of talc and hydroxyapatite. c. hydroxyapatite-based samples. The magnification in the upper right corner of (c) highlights potential diffraction peaks associated with fluorapatite (Giustetto *et al* 2013: 4255).

These discoveries indicate the utilization of a blend of two white constituents, specifically talc and bone ash (calcined hydroxyapatite), in the majority of instances. Furthermore, minor amounts of impurities were detected alongside the white pigments in nearly all examined samples (Giustetto *et al* 2013: 4254). The intermittent presence of chlorite [$(Mg,Al)_6(Si,Al)_4O_{10}(OH)_8$], often found in association with talc in natural formations, was sporadically noted (CdA7) and attributed to impurities within the filling material itself. Peaks corresponding to quartz, micas (biotite and muscovite), and feldspars (orthoclase and plagioclase), identified in most samples, may be ascribed to potential contamination from the ceramic substrate during the sampling process (Giustetto *et al* 2013: 4255).

The study by Giustetto *et al* (2013) adopted a different methodology from Scarcella and Scarcella *et al* (2011) in their research on white infills. Giustetto *et al* (2013) used Raman spectroscopy, FTIR and XRPD, whereas Scarcella *et al* (2011) employed polarised light microscopy, SEM-EDS and XRD. Raman spectroscopy seems to be less commonly used than other analytical techniques for the scientific study of ceramic decorations. In the study by Giustetto *et al* (2013), it was not very efficient, but it has been effectively used in numerous studies, as described by Bersani and Lottici (2016). The use of FTIR proved useful in the study by Giustetto *et al* (2013) for identifying minerals present within the white infills. FTIR, which is also able to detect organic materials, has also been a staple in pigment analysis and has been employed in numerous studies, including those by Franquelo *et al* (2009) and Fengwei *et al* (2017). Additionally, mineral identification was successfully aided by XRPD in the research by Giustetto *et al* (2013). Their study could have been further complemented by the use of SEM-EDS, which would have provided microphotographs of the material and EDS data on the elements present, resulting in a better understanding of the sample morphology and the elements involved.

The remainder of the section will review scientific studies conducted on white-infilled decorations on pottery from comparable timeframes but from other regions, since local studies are limited. XRPD, FTIR and X-ray Fluorescence (XRF) spectroscopy were employed in a study on white infills on Early Neolithic pottery samples originating from Pločnik, Serbia (**Figure 18**) (Perišić *et al* 2016).
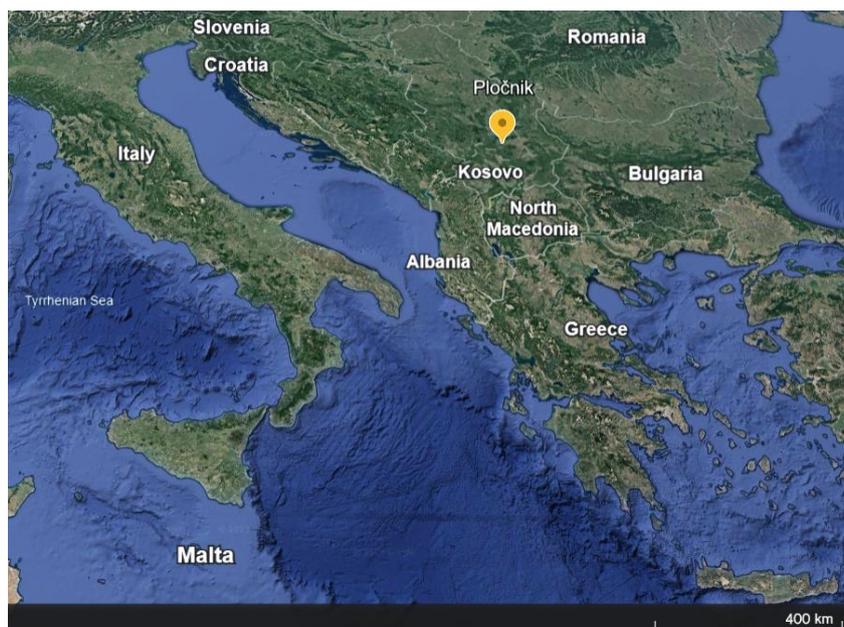
Figure 18: Map showing the location of Pločnik (Google Earth 2023)

These samples were attributed to the Vinča culture, which thrived in the central Balkan region (5500-4500 BC) (Perišić *et al* 2016: 1415). By examining the mineralogical composition of the pottery and contrasting it with locally heated clay in a modern furnace, the researchers established firing temperatures between 600 and 800 °C (Perišić *et al* 2016: 1415). Additionally, the study determined two distinct compositions for the white infills: calcium carbonate and crushed bone (Perišić *et al* 2016: 1415). This finding marked the first evidence of bones being used for decorations in the pottery of the Vinča culture at the Pločnik site (Perišić *et al* 2016: 1415).

The XRPD patterns of the samples under investigation primarily displayed quartz peaks (Perišić *et al* 2016: 1417). In addition to quartz, distinctive reflections associated with feldspars were consistently detected in all the samples (Perišić *et al* 2016: 1417). A comparison between the XRPD patterns of the pottery sherds and a raw clay sample led to the conclusion that their mineralogical compositions were highly similar (Perišić *et al* 2016: 1417). The limited amount of available material for analysis, owing to "the nature of the samples", constrained the possibility of employing more precise mineralogical determination methods that require destructive testing (Perišić *et al* 2016: 1417). Nonetheless, archaeometric studies suggest that Neolithic potters in the Balkan region likely utilised local clay for their pottery production (Spataro 2007: 147). The methodology employed by Perišić *et al* (2016) differed from that of Scarcella (2011) and Giustetto *et al* (2013). While they also utilised XRPD and FTIR like Giustetto *et al* (2013), they further incorporated the use of XRF, which offers greater analytical

sensitivity, potentially shedding light on trace elements. However, Perišić *et al* (2016) opted not to utilise SEM in their study. Its inclusion could have enriched the analysis by providing microphotographs, offering visual insights into the sample morphology

The detection of illite/muscovite in all examined pottery samples showed that the firing temperature did not exceed 900 °C, as phyllosilicates decompose between 900 and 950 °C (Perišić *et al* 2016: 1417). This contrasts with other studies on Vinča pottery from Selevac (1000 °C) (Kaiser 1990: 255), Romania (700 °C, with some samples at 850-900 °C) (Goleanu *et al* 2005: 948), Serbia (800 °C for most pottery, with some samples at 750-850 °C) (Maniatis and Tite 1981: 61) and Karanovo, Bulgaria (750 °C or lower) (Kingery and Frierman 1974: 205). **Figure 19** shows the location of the sites mentioned above.
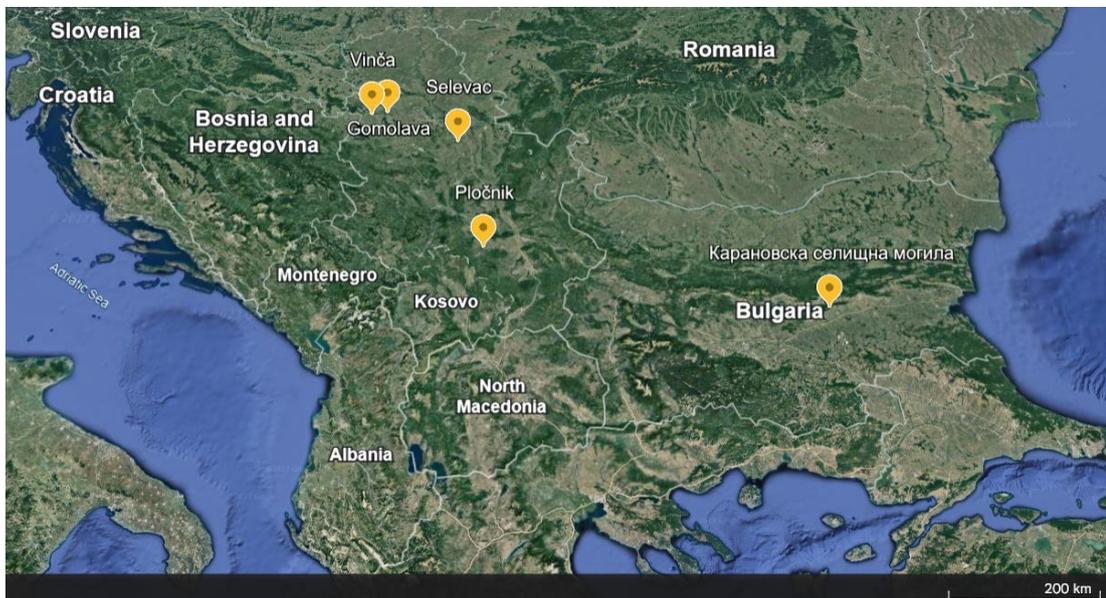
Figure 19: Map showing the geographic location of the sites of Selevac, Vinča, Pločnik, Gomolava and Karanova (Google Earth 2023)

FTIR analysis on the white infill by Perišić *et al* (2016: 1420) revealed the presence of CO_3^{2-} and PO_4^{3-} bands in sample P-9 (**Figure 20**), indicating the presence of hydroxyapatite and carbonate-hydroxyapatite, minerals commonly found in bones.
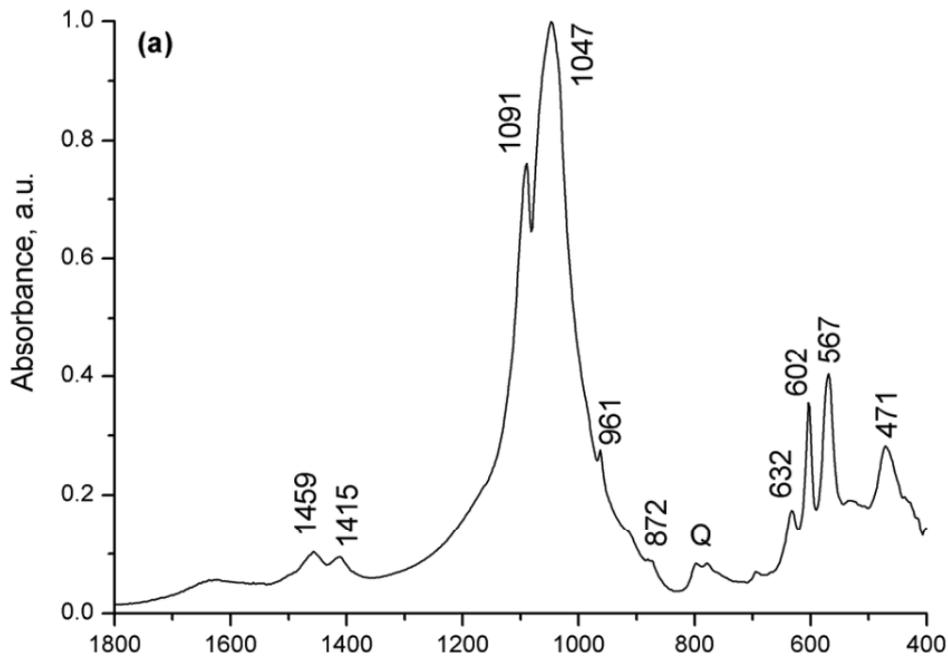
Figure 20: FTIR Spectrum of sample P-9 (Perišić *et al* 2016: 1421)

The PO_4^{3-} bands suggest that the bones underwent high-temperature processing, likely exceeding 600 °C (Perišić *et al* 2016: 1420). Additionally, the presence of a distinctive band at 632 cm^{-1} in the FTIR spectrum further confirms high-temperature calcination, as this band is associated with the vibrational mode of hydroxyl groups within hydroxyapatite and emerges at elevated temperatures during calcination (Odriozola *et al* 2007: 1800). These results strongly indicate that the bones utilised in the manufacturing of the white infill decorations in sample P-9 were calcined at high temperatures ranging from 700 to 900 °C (Perišić *et al* 2016: 1421). Although it is not possible to definitively determine whether the bone material was independently fired or together with the vessel, it is likely that Vinča potters calcined the bones before crushing them to create the white infill, as burned bones are easier to process into fine powder (Perišić *et al* 2016: 1421). As a result, it can be deduced that the pigment known as "Bone White," obtained from the calcination of animal bones, teeth and/or antlers, was used for the purpose of white decoration on sample P-9 (Perišić *et al* 2016: 1422). These findings indicate that Vinča culture potters were knowledgeable in the technique of incorporating calcined bones to achieve white decorations on pottery (Perišić *et al* 2016: 1422).

In contrast, another FTIR spectrum of the white pigment from sample P-10 (**Figure 21**) displayed an "intensive and broad band" related to calcium carbonate ($CaCO_3$), the primary constituent of this white decorative infill (Perišić *et al* 2016: 1422).
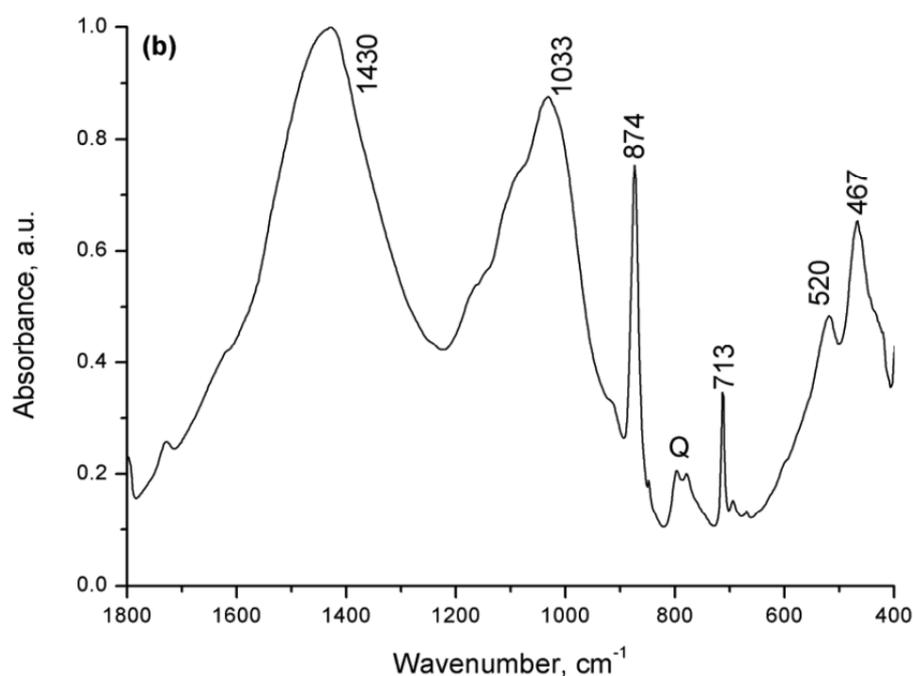
Figure 21: FTIR Spectrum of sample P-10 (Perišić *et al* 2016: 1421)

The characteristics of this band suggest that $CaCO_3$ was added after the firing of the pottery as a subsequent production step (Perišić *et al* 2016: 1422). This is because when $CaCO_3$ is subjected to high temperatures, significant alterations in its FTIR bands occur. Specifically, these changes involve a shift towards lower wavenumbers in the spectrum (Xu and Poduska: 2014: 17634). Furthermore, the characteristic v4 band at 713 cm^{-1} undergoes broadening, a decrease in intensity, and ultimately diminishes almost entirely, particularly when exposed to temperatures exceeding 600 °C (Xu and Poduska 2014: 17635). This aligns with visual observations of the sample, which implied the addition of pigments during a later stage of production (Perišić *et al* 2016: 1422). In the Energy Dispersive X-Ray Fluorescence (EDXRF) spectra of white infills from samples P-9 and P-10, signals corresponding to calcium were also observed, consistent with the results derived from the FTIR analysis (Perišić *et al* 2016: 1422). Lastly, the presence of the characteristic doublet of quartz in both FTIR spectra, as well as the Si-O band in sample p-10, located at 1033 cm^{-1}, is likely due to the scraping of the ceramic body during sampling (Perišić *et al* 2016: 1422). Nevertheless, it is important to acknowledge the possibility of impurities in the infill mixture (Perišić *et al* 2016: 1422).

In another study, Pirovska (2021) conducted scientific analysis of pigments utilised in two types of pottery decoration namely, paint (**Figure 22**) and infills (**Figure 23**). The study encompassed 60 pottery sherds retrieved from 13 archaeological sites across Western, Central and Eastern Bulgaria, dating back to the Neolithic and Chalcolithic periods (**Figure 24**)

(Pirovska 2021: 19). The employed methodology heavily relied on XRD, given that the pigments studied consisted predominantly of minerals.
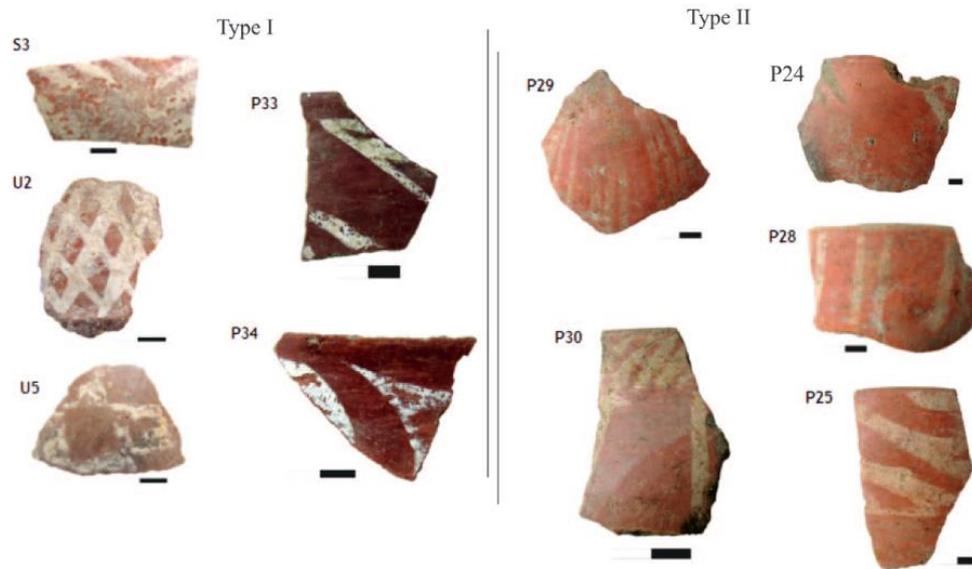
Figure 22: Pottery fragments featuring white pigment employed as paint (Pirovska 2021: 22)
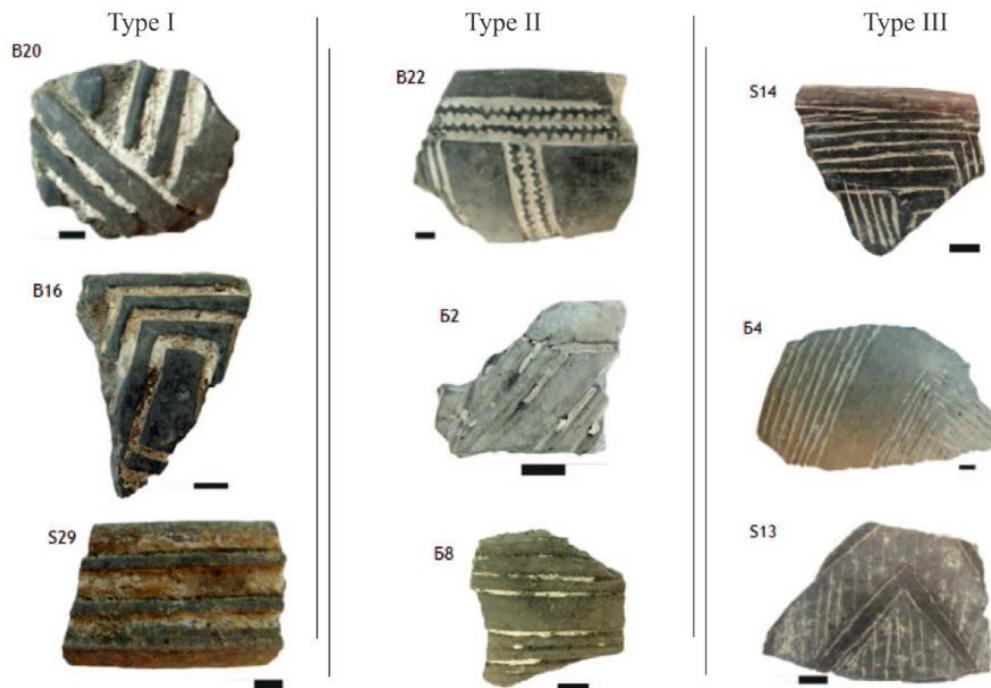
Figure 23: Pottery fragments featuring white pigment employed as infill (Pirovska 2021: 24)
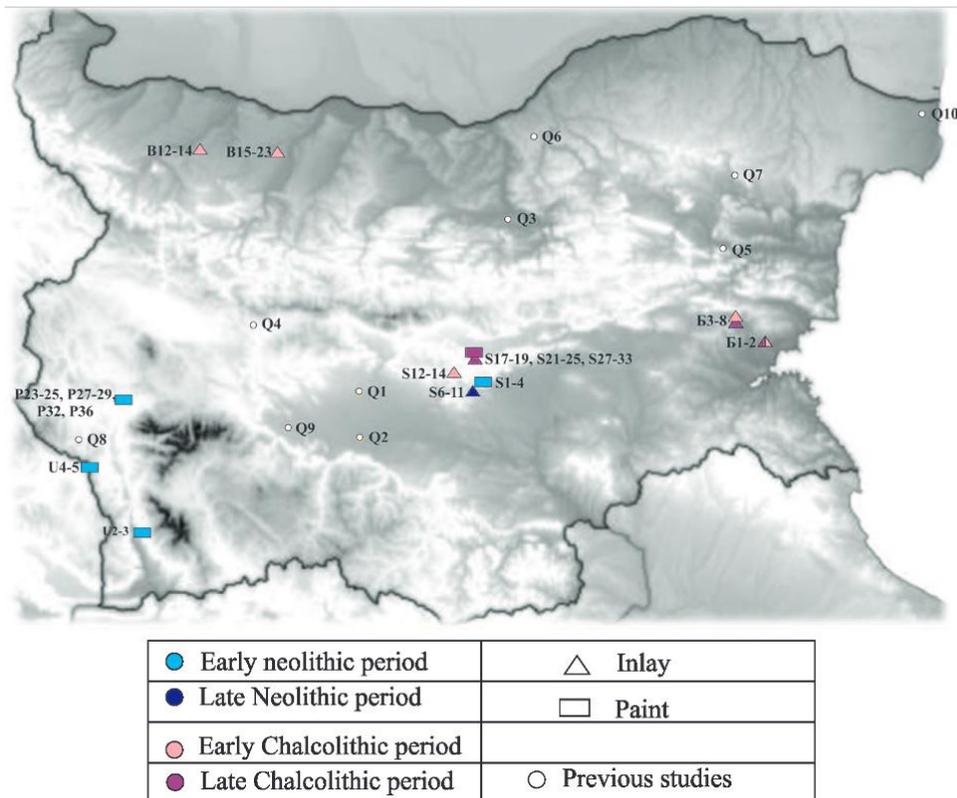
Figure 24: Geographical representation of Bulgaria indicating the specific sampling site locations along with corresponding sample numbers (Pirovska 2021: 19)

The samples selected for analysis originated from archaeological sites spanning the territories of diverse cultural groups (Pirovska 2021: 24). In Southwest Bulgaria, particularly during the Neolithic period, the West-Bulgarian painted ceramics culture thrived as part of the broader Balkan-Anatolian Early Neolithic Complex (5th millennium). Characterised by distinctive, white-painted ceramic decorations, this cultural group was exemplified by 12 pottery fragments retrieved from three archaeological sites (P and U in **Figure 24**). XRD of the white material revealed predominant use of calcite, as detailed in **Table 5**, and a sole sample showing gypsum. Moving to Central Bulgaria, specifically within the Thracian Lowland during the Neolithic period, another distinct cultural group emerged. This group was represented by six pottery fragments obtained from two archaeological sites (S in **Figure 24**). The analyses conducted revealed calcite as the predominant mineral in the composition of the white paint (**Table 5**). Additionally, talc was identified as an additive in two of the samples. Local variations in mineral impurities were also discerned, such as gypsum in the Pernik region of Northwest Bulgaria and talc in the vicinity of Stara Zagora, Central Bulgaria. Within the confines of this early Neolithic settlement, two distinct types of ceramic decoration paint coexisted, which are visually indistinguishable. This prompted an inquiry into whether these

materials were imported from an external source or if, within the same village, two distinct methods were utilised to produce the white material.

№ по ред	проба	Q	illite	Fs	calcite	talc	gypsum
1	S1	~	~	-	+	+	-
2	S2	+	-	-	+	-	-
3	S3	+	-	~	+	+	-
4	S4	+	-	~	+	-	-
5	S25	~	-	~	+	-	-
6	S28	+	-	-	+	-	-
7	P23	+	+ Ms	+ Pl	-	-	-
8	P24	+	+ Ms	+	-	-	-
9	P25	+	+ Ms	+ Pl	~	-	-
10	P27	+	+	+	+	-	-
11	P28	+	+	+ Pl	-	-	-
12	P29	+	+ Ms sericite	+ Pl	-	-	-
13	P32	-	+ Ms	-	+	-	-
14	P36	+	+ Ms sericite	+ Pl	+	-	+
15	U2	+	+	+	+	-	-
16	U3	+	+	+	+	-	-
17	U4	+	+	-	~	-	-
18	U5	+	+	+	-	-	-

Table 5: Mineralogical composition of painted samples analysed through XRD (Pirovska 2021: 25)

During the early Chalcolithic period in Bulgaria (*c.* 4900-3800 cal. BC) (Boyadzhiev 2014: 163), an emergence of white paste infill decoration occurred (Pirovska 2021: 24). Concurrently, the Gradeshnitsa cultural group flourished in Northwestern Bulgaria. 12 pottery fragments from two archaeological sites were analysed in this study (B in **Figure 24**). XRD revealed that the white paste used for these infilled fragments was predominantly composed of calcite (**Table 6**). Three samples from the village of Brenitsa, Vratsa, exhibited an alternative mineral composition devoid of calcite. Two of these were composed of gypsum and barite or bassanite. The presence of barite deposits in the geographic vicinity of the archaeological site, suggest the utilisation of local raw materials for pigment production (Pirovska 2021: 25). The other sample was composed of apatite.

№ по ред	проба	Q	illite	Fs	calcite	clinochlore	kaolinite	dolomite	gypsum	bassanite	barite/ hyalophane	apatite
1	S6	+	-	~	+	-	-	-	-	-	-	-
2	S7	+	-	+ Pl	+	-	-	-	-	-	-	-
3	S8	+	-	+	+	-	-	-	-	-	-	-
4	S9	+	+	~	+	-	-	-	-	-	-	-
5	S10	+	+	-	+	-	-	-	-	-	-	-
6	S11	+	-	-	+	-	-	-	-	-	-	-
7	S12	+	-	-	+	-	-	-	-	-	-	-
8	S13	+	-	-	+	+	-	-	-	-	-	-
9	S14	+	+	-	-	+	-	-	-	-	-	-
10	S17	+	-	+ Pl	+	-	-	-	-	-	-	-
11	S18	+	-	~ Px	+	-	-	-	-	-	-	-
12	S19	+	+	+	-	-	-	-	-	-	-	-
13	S21	+	-	-	+	-	-	-	-	-	-	-
14	S22	+	+	+ Pl	+	-	-	~	-	-	-	-
15	S23	+	-	~	+	-	-	-	-	-	-	-
16	S24	+	-	~	+	-	-	-	-	-	-	-
17	S27	+	-	~	+	-	-	-	-	-	-	-
18	S29	+	~	~	+	-	-	-	-	-	-	-
19	S30	+	+	~	+	-	-	-	-	-	-	-
20	S31	+	+	-	+	-	-	+	-	-	-	-
21	S32	+	-	~	+	-	-	-	-	-	-	-
22	S33	+	-	~	+	-	-	-	-	-	-	-
23	B12	+	-	~	+	-	-	-	-	-	-	-
24	B13	-	-	-	+	-	-	-	-	-	-	-
25	B14	~	-	-	+	-	-	-	-	-	-	-
26	B15	~	-	-	+	-	-	-	-	-	-	-
27	B16	+	+ Ms	+ Px	+	-	~	-	-	-	-	-
28	B17	~	-	-	+	-	-	-	-	-	-	-
29	B18	~	-	~	+	-	-	-	-	-	-	-
30	B19	-	-	-	-	-	-	-	+	+	-	-
31	B20	-	-	-	+	-	-	-	-	-	-	-
32	B21	-	-	-	+	-	-	-	-	-	-	-
33	B22	-	-	-	-	-	-	-	+	-	+	-
34	B23	+	-	-	-	-	-	-	-	-	-	+
35	Bu1	+	+ Ms	-	-	-	-	-	+	-	-	-
36	Bu2	+	-	-	+	-	-	-	-	-	-	-
37	Bu3	+	-	-	+	-	-	-	+	-	-	-
38	Bu4	+	+	+	+	-	-	-	-	-	-	-
39	Bu5	+	-	+	+	-	-	+	-	-	-	-
40	Bu6	+	+	+	+	-	-	-	-	-	-	-
41	Bu7	+	-	-	+	-	-	-	+	-	-	-
42	Bu8	+	-	-	+	-	-	+	-	-	-	-

Table 6: Mineralogical composition of infilled samples analysed through XRD (Pirovska 2021: 25)

Twenty-two pottery fragments obtained from three archaeological sites (S in **Figure 24** and **Table 6)** were used in the study to represent the cultural groups in Central Bulgaria during the Chalcolithic period (Pirovska 2021: 25). According to XRD analysis, calcite was identified as the primary mineral used in the production of the white paste infill. One sample exhibited the presence of dolomite. The author suggests that a singular deposit could have served as the primary source for extracting the raw materials used in white paste production throughout the settlement's entire occupation. In another archaeological site within the same micro-region, XRD analysis of the white infill used for three pottery fragments also identified calcite as the major mineral. However, in two of the fragments, clinochlore was also detected, indicating a distinct composition of the utilised production materials. This observation suggested that the white infill was likely locally produced, with the raw material extracted from a different deposit.

During this time period, a distinct cultural group emerged in the eastern part of the Thracian Plain during the Chalcolithic period (Pirovska 2021: 27). This tradition was represented by eight pottery fragments from two archaeological sites in this study (Bu in **Figure 24**). Similar to the findings in Central Bulgaria, calcite was identified as the primary component in all samples, except for one (refer to **Table 6**). XRD also revealed variations in the composition of the white paste, with the identification of gypsum in three sherds and dolomite in two fragments. The presence of such diverse raw materials suggested the utilisation of two distinct deposits, indicating the presence of two separate pottery production centres. This is further supported by the clear differentiation observed in the XRD analysis, which distinguished between two types of main compositions used for the production of white infill, namely, pigments with calcite and those without (**Table 6**). Distinct impurities in the white infill compositions were attributed to local features. For example, dolomite and clinochlore in the regions of Stara Zagora (Central Bulgaria) and Burgas (Central East Bulgaria), and kaolinite in Stara Zagora (Central Bulgaria) and Vratsa (Northwest Bulgaria). Certain mineral impurities, such as bassanite, barite or hyalophane, can be identified as local features, potentially linked to the selection of diverse material sources (Pirovska 2021: 28). The diversity in white infill composition within a small micro-region could signify heightened mobility of people and goods. However, a more comprehensive investigation is essential to validate this conclusion. This study suggested that, from the Neolithic to the Chalcolithic period in Bulgaria, no discernible alteration in the manufacture of the white infill was identified (Pirovska 2021: 29).

The investigation of white-infilled pottery decorations has garnered attention in archaeological and scientific research since at least the 1980s when Gâță and Mateescu (1987) studied an assemblage of pottery fragments from two Romanian Chalcolithic sites (5th millennium BC), Vădastra and Crușovu (**Figure 25**), which are located approximately 8 kilometres apart.
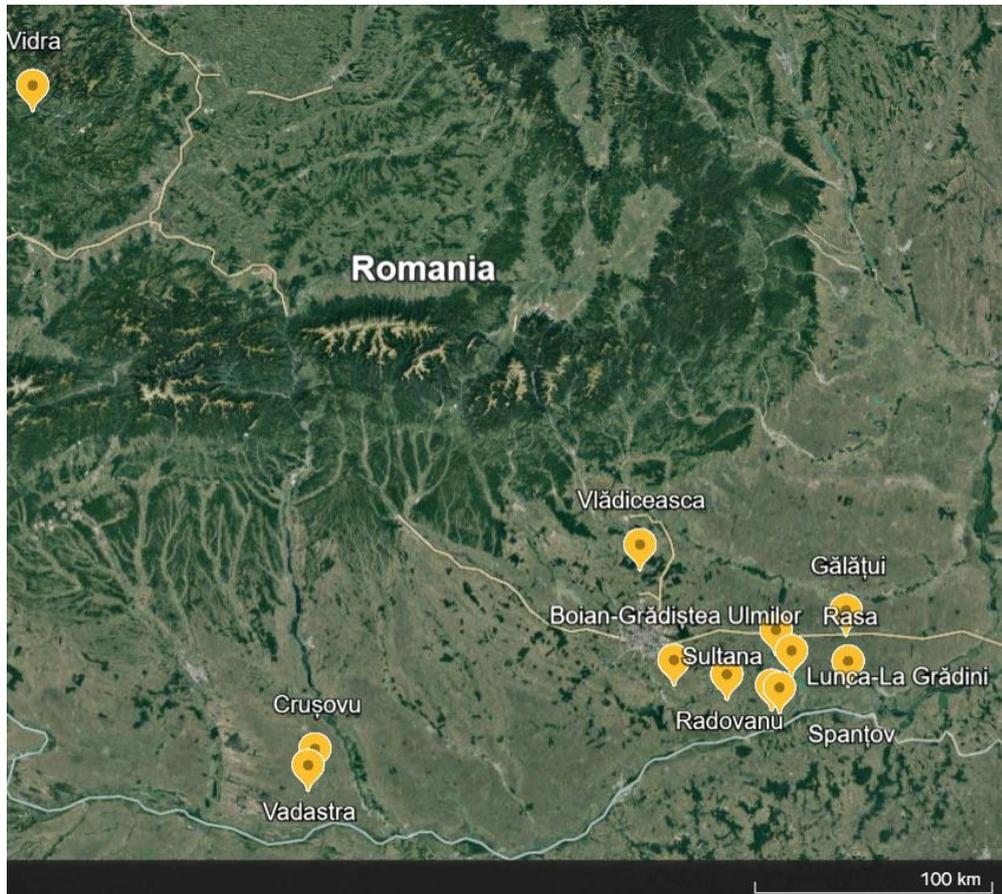
Figure 25: The archaeological sites mentioned in the studies conducted on the white pigments used in Romanian pottery. Vădastra and Crușovu in Gâță and Mateescu (1987), Gălățui-Movila Berzei, Boian-Grădiștea Ulmilor, Grădiștea Călărași, Lunca-La Grădini and Rasa in Niculescu (2003), Spantov in Dragoman *et al* (2019) and Opriș *et al* (2019), Radovanu-La Muscalu in Ștefan (2019) and Sultana, Vidra and Vlădiceasca in Opriș *et al* (2022)

This initial study, focusing on the composition and application of white pigments used as paint and incrustation (**Figure 26**), paved the way for an extensive exploration of this practice in Romania. Gâță and Mateescu were some of the first to analyse white-infilled decorations from Romania. Their methodology employed Infra-Red absorption (IR) and XRD. Their methodology laid the groundwork for further exploration of this practice. However, a broader

range of techniques could have offered a more comprehensive understanding of the infill's composition and potential formation processes.

Through their investigation, Gâţă and Mateescu identified the presence of quartz (SiO_2), calcite, illite and kaolinite, feldspars, iron oxides, manganese and aluminium within the white material (1987: 217).
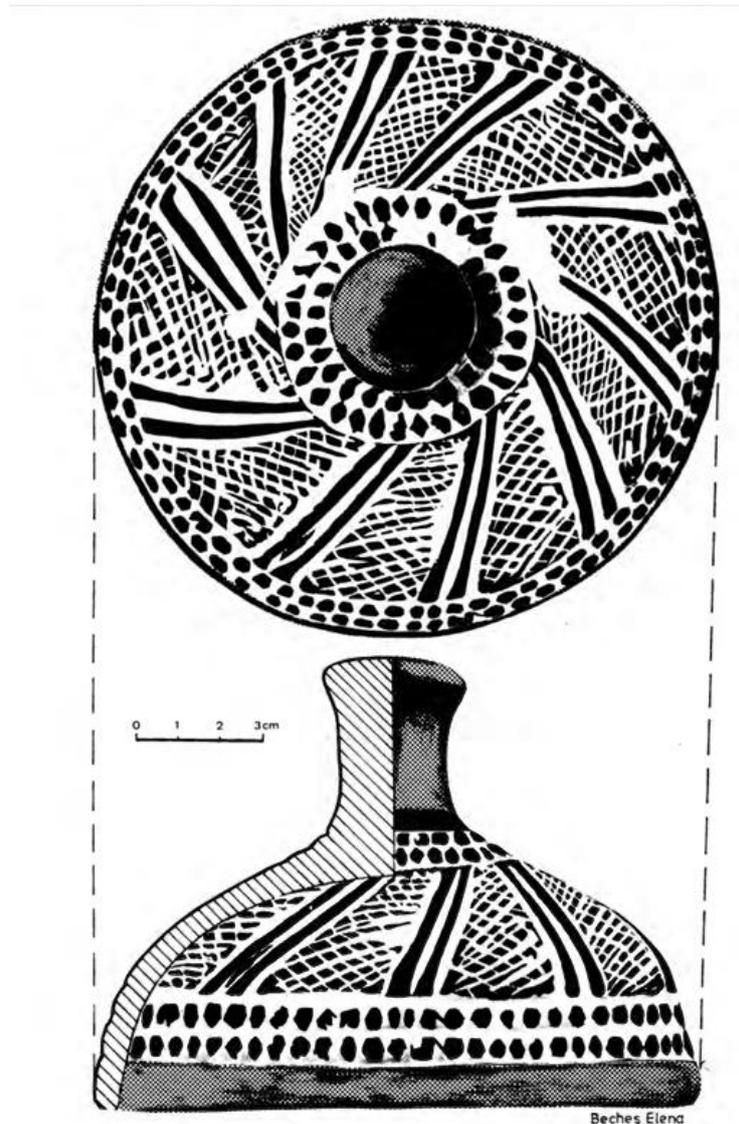
Figure 26: Lid of a Vădastra II phase (5th millennium BC) ornamental pot, featuring white-infilled decorations. Red coloration was applied to the rim and shoulder (Gâţă and Mateescu 1987: 203)

They also determined, through the chemical analysis of local sources, that the carbonate neoformations within the vicinity of both settlements served as the primary source of the white material (Gâţă and Mateescu 1987: 217). This conclusion was reached by mineralogically analysing samples from deposits in the vicinity of the Vadastra site using XRD. The analysis

showed that these samples had a distribution of calcite and quartz similar to that of the white infills (Gâță and Mateescu 1987: 208). However, a potential issue arises from the assumption that the Vadastra people exclusively used these local sources. While it is logical to infer that they would utilise readily available materials, other sources with similar distributions of calcite and quartz might have also been used. Additionally, Gâță and Mateescu put forth the proposition that local raw materials were carefully selected and processed to maximize the quantity of calcite while minimizing the level of quartz, thus achieving the desired white pigment (Gâță and Mateescu 1987: 208). The disparity in quartz content between the raw materials sourced from Crușovu and Vădastra was further reflected in the higher quartz ratio found in the white substance utilised to decorate the pottery from Crușovu, as compared to the pottery from Vădastra (Gâță and Mateescu 1987: 210).

A study by Niculescu (2003) focused on the white infill used to decorate Boian pottery during the Giulești phase (5th millennium BC), which coincided with the Vădastra period. Sixteen pottery samples, collected from five Boian sites in Călărași County, were analysed using XRD. XRD has been already noted as an effective technique for identifying the mineral phases present in a material. Ideally, this method should be complemented by microscopic and elemental analysis techniques, such as SEM-EDS, for a more comprehensive understanding of the sample composition. Their analysis demonstrated that the predominant component of the white infill was calcium carbonate (Niculescu 2003: 113). In certain instances, this was found alongside quartz, with the latter being linked to gismondine and diopside. However, at Gălățui-Movila Berzei, the analysis revealed the combination of bone with calcite in one sample, while another sample exclusively comprised hydroxyapatite.

Two later studies concentrated on investigating the white paste employed for decorating Boian pottery during the late Spanțov phase (4800-4500 BC) (**Figure 25**) (Dragoman *et al* 2019, Opriș *et al* 2019). Particle-induced X-ray emission (PIXE) and XRD were utilised in these studies. PIXE (Particle Induced X-ray Emission) is a non-destructive analytical technique for elemental analysis, generally offering superior sensitivity compared to other techniques like EDS, meaning it can detect and quantify trace elements with greater precision and accuracy. However, EDS seems to enjoy wider use for analysing white-infilled decorations, most probably, due to its integration with SEM, allowing for simultaneous elemental analysis and morphological characterisation of the decoration material, alongside PIXE's limited beamtime opportunities. PIXE, although less accessible than SEM-EDS, has also proven valuable in scientific studies of pigments (Turos *et al* 2009, Beck *et al* 2012). The use of PIXE and FTIR

techniques in these studies revealed that the white paste compositions consisted of calcium compounds, different types of quartz, or high levels of aluminium-silicates due to the inclusion of kaolinite in the mixture (Dragoman *et al* 2019: 81, Opriş *et al* 2019: 92). In the case of another study focusing on a sample from Radovanu-La Muscalu, a site corresponding to the Spanţov phase, it was revealed that the white infill mainly consisted of calcite, with a lower concentration of quartz (Ştefan 2019: 152).

More recently, Opriş *et al* (2022) analysed the white-infilled decorations of thirty-one pottery samples (**Figure 27**) obtained from the sites of Sultana, Vidra and Vlădiceasca (4900-4600 BC) (**Figure 25**). The study utilised a number of archaeometric techniques including in-air PIXE, EDX, XRD, FTIR, SEM and Electron paramagnetic resonance (EPR). EPR was used for the analysis of the paramagnetic centres. This tool can detect paramagnetism in certain materials that are weakly attracted by an applied magnetic field. Sample preparation was carried out in the following manner. The upper pigment layer, often mixed with dirt, was gently scraped and removed using a metal spatula. Then, in-air PIXE was conducted on the white infill present on the pottery sherds. For XRD, FTIR, SEM-EDX, and EPR analyses, infill samples were collected using a metal needle after the initial layer of infill mixed with dirt had been removed (Opriş *et al* 2022: 3). This process abstained from using solvents or water to maintain the original composition intact. Additionally, the study prepared reference materials (Opris *et al* 2022: 3), which comprised calcium-based materials obtained from the underlying loess layer at the Sultana-Gheţarie site, along with animal bone samples from a Boian-Vidra pit within the Chalcolithic cemetery area at the same site (Opriş *et al* 2012). Before analysis, the animal bones underwent a three-hour calcination process at 800°C to yield bone ash, while the calcium-based materials underwent firing at 700°C for 15 minutes, considering the thermal decomposition of calcite above 750°C.

The methodology employed by Opriş *et al* (2022) offered a strong foundation for analysing the white infill decorations on Boian pottery. Firstly, their use of several established and specialised techniques (XRD, FTIR, SEM-EDX PIXE, EPR), contrary to most previous studies which only adopted one to two techniques, provides a comprehensive understanding of the infill's composition, mineralogy and potential formation processes. Additionally, the inclusion of reference materials from potential source materials (loess and bone) strengthened the analysis by facilitating comparison and identification of the infill's origin and composition.
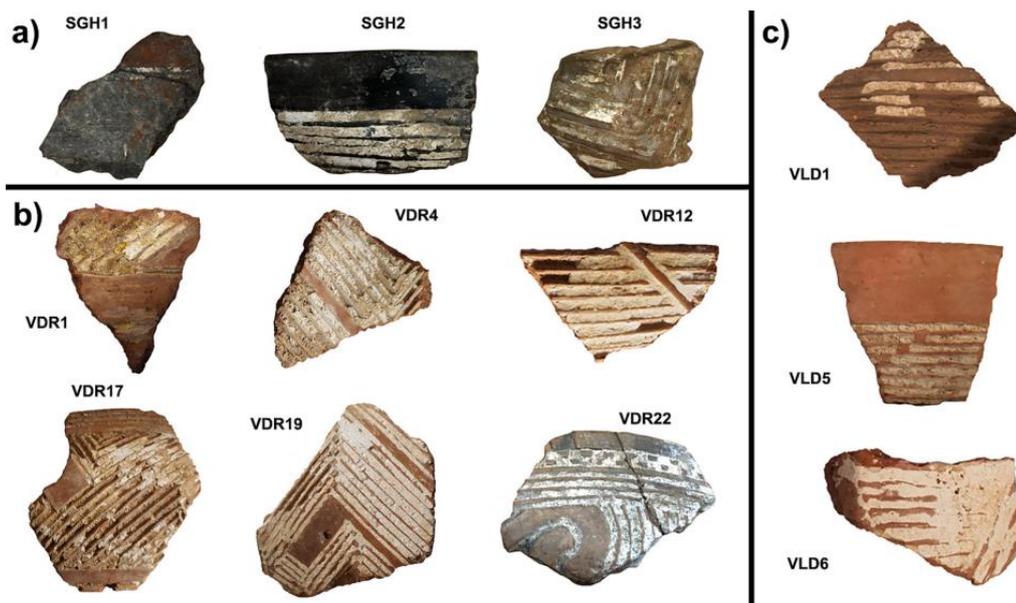
Figure 27: Some pottery sherds showing white-infilled decorations analysed in Opriș et al (2022) from (a), Vidra (b) and Vlădiceasca (c)

Opriș *et al* (2022) determined, through their analysis, the predominantly inorganic composition of the investigated white infills, consisting primarily of calcite, bone or combinations thereof, occasionally with elevated levels of silica-rich sediments (Opriș *et al* 2022: 8). Mineralogical assessments confirmed the prevalence of calcite in nearly all the white infill samples, except for sample VLD5 (**Table 7**). Bone material was identified in two samples (VLD4 and VLD6) and was observed in conjunction with calcite and sporadically with silicon-based compounds in 11 other samples. Based on their compositions, the analysed infills were classified into four main groups: a) infills primarily composed of calcite occasionally intermixed with minor quantities of silicates (16 specimens); b) combinations of calcite (dominant) and bone (minor) in various proportions, typically with traces of silicates (11 specimens); c) bone (predominant) infills with minimal amounts of calcite and quartz; and d) silica-rich white infills (VLD2 and VLD5).

Sample	Calcite		Quartz from XRD	Hydroxyapatite (HA) from XRD	HA-Crystallinity degree from FTIR spectra)
	from XRD	Mn signal from EPR			
VDR1	Major	Very weak	Very weak	Minor	2.7
VDR2	Dominant	Minor	Very weak	Weak	4.9
VDR3	Dominant	Dominant	Not detected	Not detected	–
VDR4	Dominant	Dominant	Not detected	Not detected	–
VDR5	Major	Very weak	Very weak	Minor	3.0
VDR6	Dominant	Dominant	Very weak	Not detected	–
VDR7	Major	Minor	Weak	Minor	3.0
VDR8	Dominant	Dominant	Very weak	Not detected	–
VDR9	Dominant	Dominant	Not detected	Not detected	–
VDR10	Dominant	Minor	Weak	Not detected	–
VDR11	Dominant	Dominant	Not detected	Not detected	–
VDR12	Major	Major	Very weak	Minor	6
VDR13	Dominant	Weak	Not detected	Very weak	2.5
VDR14	Dominant	Weak	Very weak	Not detected	–
VDR15	Dominant	Dominant	Weak	Not detected	–
VDR16	Dominant	Weak	Very weak	Not detected	–
VDR17	Dominant	Weak	Weak	Not detected	–
VDR18	Dominant	Dominant	Very weak	Not detected	–
VDR19	Dominant	Dominant	Weak	Very weak	4.8
VDR20	Dominant	Major	Yes	Very weak	3.7
VDR21	Dominant	Weak	Yes	Not detected	–
VDR22	Dominant	Dominant	Very weak	Very weak	5.5
VLD1	Dominant	Major	Weak	Very weak	4.0
VLD2	Very weak	Not detected	Dominant	Not detected	–
VLD3	Dominant	Not detected	Weak	Weak	5.2
VLD4	Very weak	Dominant	Weak	Dominant	8.1
VLD5	Not detected	Not detected	Dominant	Not detected	–
VLD6	Weak	Not detected	Weak	Dominant	8
SGH1	Dominant	Dominant	Very weak	Not detected	–
SGH2	Dominant	Dominant	Weak	Not detected	–
SGH3	Dominant	Very weak	Weak	Not detected	–

Table 7: Mineralogical qualitative data of the analysed white infills in Opriş *et al* (2022)

The authors discussed possible sources for the materials used in the white infills. They suggested that calcite, prevalent in most analysed samples, could have been sourced from geological layers such as limestone, chalk, marble or marl, as well as carbonate neo-formations and shells of terrestrial and water molluscs (Opriş *et al* 2022: 10). While distant calcite sources were available, the local carbonate concretions in loess and shells of molluscs were more accessible to prehistoric communities near the respective archaeological sites. They conducted a comparative experiment using XRD, FTIR, and SEM-EDX to establish similarities between carbonate concretions found near the archaeological sites and the white-infilled decorations obtained from these sites. This experiment confirmed that the carbonate concretions derived from the local loess were the probable sources of calcite used in the white-infilled decorations.

Investigating the origin of hydroxyapatite in the white infills involved comparing pigments containing significant amounts of hydroxyapatite (VLD4 and VLD6) with bone ash reference material (Opriş *et al* 2022: 10). Results from XRD, FTIR and SEM analyses showed similarities, consistent with previous studies on the source of hydroxyapatite in ancient pottery pigments (Odriozola and Hurtado Pérez 2007, Giustetto *et al* 2013). These results strongly suggested that bone ash was employed as a whitening agent in certain Boian-Vidra infills (Opriş *et al* 2022: 10). Additionally, the authors aimed to identify the animal species contributing to the hydroxyapatite in the white infills using SEM. For each sample (**Figure 28**: a-h) and reference samples (**Figure 28**: i-p), SEM created four photographs at different magnification levels (5000x, 10000x, 20000x and 50000x) (Opriş *et al* 2022: 6). This

facilitated examination and comparisons particles and matrices. The anticipation was that specimens with similar chemical and mineral make-ups would exhibit comparable microstructures. The distinctive grains and texture of hydroxyapatite observed under SEM, consistent with findings in the literature (Roberts *et al* 2008, Doostmohammadi *et al* 2012) and the reference bone ash of sheep and cattle (**Figure 28**: m-p), were evident in two Vlădiceasca specimens (VLD4 and VLD6) (**Figure 28**: e-f).
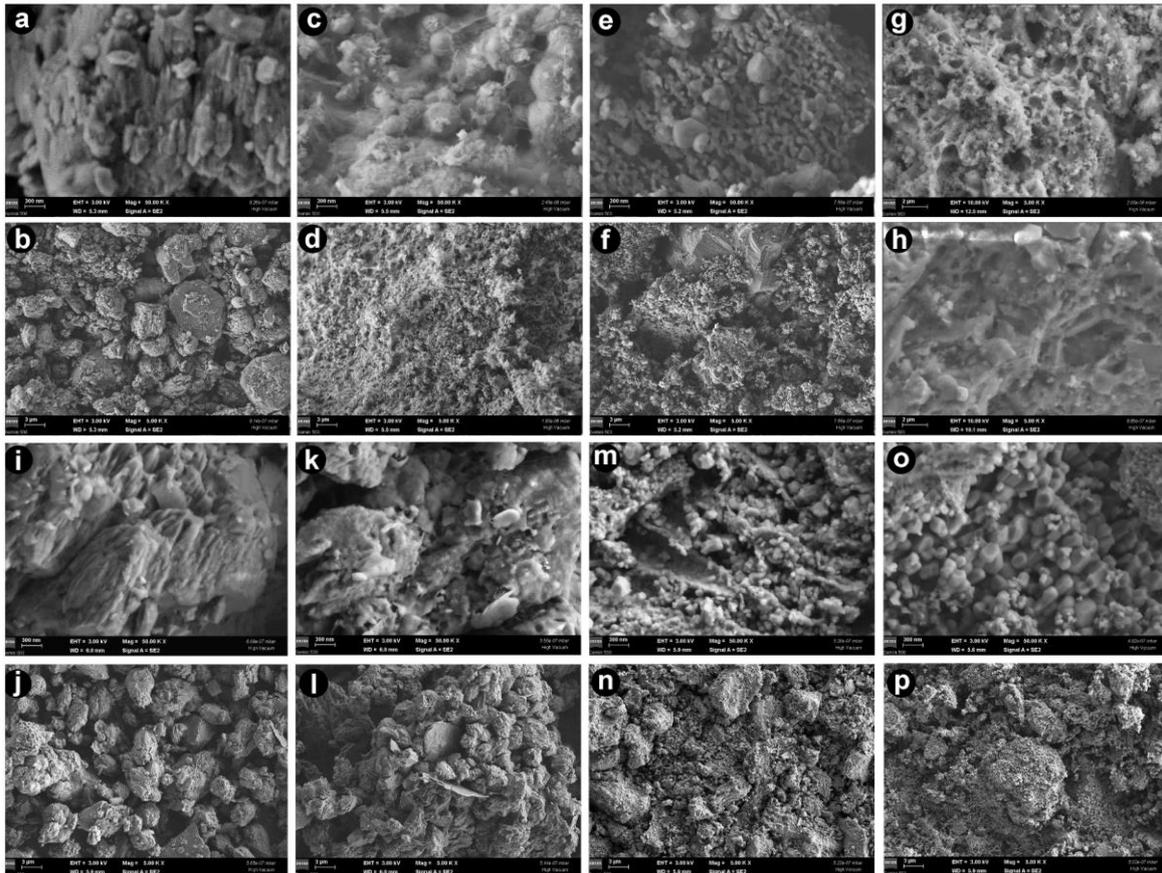
Figure 28: SEM images showing pigment samples and reference materials: SGH2 with mainly calcite (a-50000x, b-5000x), VDR1 exhibiting a mixture of calcite and hydroxyapatite (c-50000x, d-5000x), VLD6 with mainly hydroxyapatite (e-50000x, f-5000x), VLD2 depicting damaged diatoms (g-5000x) and VLD5 also displaying the damaged skeletal structures of diatoms (h-5000x). Additionally, the reference materials included unburnt calcium-based minerals (i-50000x, j-5000x), fired carbonate concretions at 700 ◦C (k-50000x, l-5000x), fired sheep bone at 800 ◦C (m-50000x, n-5000x) and fired bovine bones at 800 ◦C (o-50000x, p-5000x) (Opriș *et al* 2022: 8).

Additionally. EPR measurements conducted on Early Chalcolithic sheep and cattle bone samples, subjected to controlled conditions at 800 °C, revealed resemblances between the hydroxyapatite in pottery sherd VLD4 (**Figure 29**) and the bone ash of sheep (**Figure 30b**) (Opriş *et al* 2022: 6).
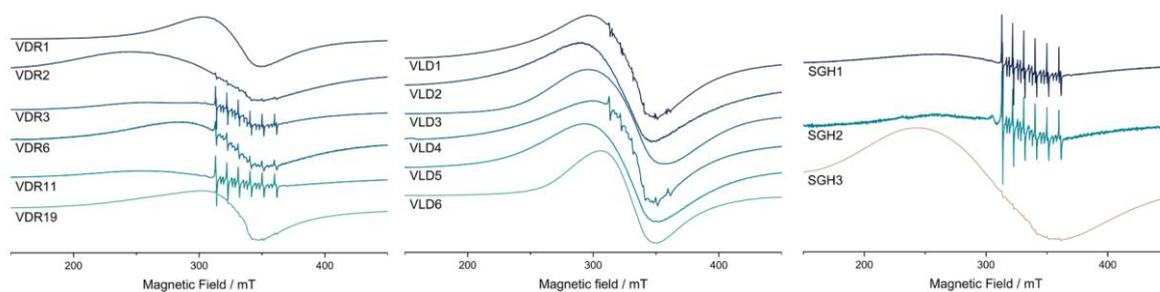
Figure 29: EPR spectra of the white infills from Vidra (left), Vlădiceasca (centre) and Sultana-Ghețarie (right) (Opriş *et al* 2022: 8)
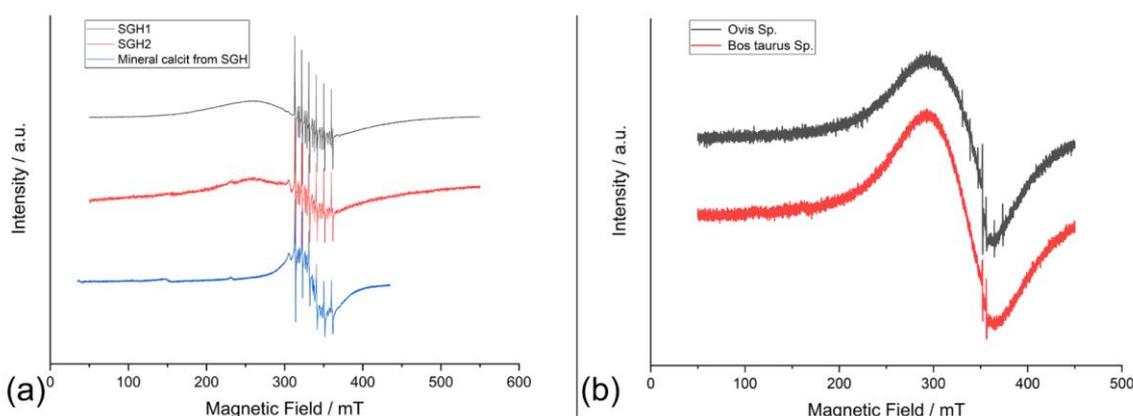
Figure 30: EPR spectra of the white infills in SGH1 and SGH2, and carbonate concretions from Sultana-Ghețarie heated at 700 °C (a). EPR spectra sheep and cattle reference bones (b) (Opriş *et al* 2022: 9)

Given the constrained number of reference materials and the significant variability in EPR spectra among infills containing hydroxyapatite, the observed resemblance may be coincidental (Opriş *et al* 2022: 11). The authors noted that further measurements involving a more diverse range of animal remains are crucial to acquire pertinent data which could be useful in identifying the species used in creating the white pigment. Furthermore, the occurrence of small quantities of silicates in most samples may have diverse origins, including, the unintentional inclusion of natural materials, the ceramic sherd itself during scraping, post-depositional build-up or accumulated storage dirt. Lastly, the prevalence of silicon-based

minerals in samples VLD2 and VLD5 suggested the utilisation of white siliceous sources. The SEM images distinctly revealed the existence of circular pores, characteristic of microscopic siliceous shells of diatoms (**Figure 28**: g-h).

Opriș *et al* (2022: 11) also discussed possible manufacturing practices used in the creation and application of the white infill. Water or an alternative liquid might have been utilised to make the paste adhesive, which was subsequently inlaid into the decorations of the vessels, either prior to, or following, the firing process. Experiments carried out by the authors involving white pastes derived from local carbonate sources and burnt animal bones revealed that ground carbonate concretions, when mixed with water, easily formed an adhesive paste, adhering to the decorations. Alternatively, mixtures of ground burnt animal bones with water did not produce a binding paste, rendering the infilling process more challenging. They inferred that organic binders, which underwent complete consumption during the firing process, were employed, for instances where bone ash was ascertained to be the predominant component.

The identification of the firing procedure posed challenges in the study due to secondary firing of the ceramics (Opriș *et al* 2022: 11). The samples were found during excavations were the primary focus was on investigating burnt structures within a Chalcolithic tell settlement. The reddish hue of the sherds served as evidence of secondary firing since Boian excised pottery, typically discovered in non-burnt contexts, is predominantly black or dark grey. This assertion was supported by two pottery sherds discovered in a refuse pit at Sultana-Ghețarie (SGH 1 and SGH 2), which exhibited a black coloration. In these cases, the white infills were composed of calcite and showed no discernible traces of firing treatment.

The provided list of archaeometric analyses concerning white-infilled decorations is not intended to be comprehensive. Instead, it reviews several scientific studies on the matter that temporally relate to the present study. The purpose is to highlight existing research on the analyses of white-infilled decorations, setting the context for the present study. It should be noted, however, that other analytical studies pertaining to the archaeometric analysis of white-infilled decorations, which have not been specifically mentioned in this research project, exist. Such studies include but are not limited to those by (Rychner-Faraggi and Wolf 2001, Sziki *et al* 2003, Curtis *et al* 2010, Lantes-Suárez *et al* 2010, Pinillos de la Granja *et al* 2022, Vico *et al* 2022). The mentioned studies employed similar methodologies to those discussed in the **Literature Review**, utilising SEM, XRD, PIXE and Raman spectroscopy as the scientific techniques for analyses. The results of the analysed white infills in these studies also exhibited

resemblances to the papers reviewed, with most of the white pigments comprising bone, calcium carbonate, talc, kaolinite and quartz as the primary minerals identified. However, certain unique aspects were identified in these papers. For example, Lantes-Suárez *et al* (2010) identified talc as the predominant mineral in the white paste, occasionally accompanied by serpentine, plagioclase or amphibole minerals. Serpentine and amphiboles were not mentioned in any of the reviewed papers, while plagioclase was only referenced in the study by Giustetto *et al* (2013), who noted its association with natural talc outcrops as a feldspar impurity. This finding aligns well with the observations of Lantes-Suárez *et al* (2010), who detected plagioclase in the analysed talc-based white-infilled decorations. Another noteworthy discovery highlighted in the studies mentioned is the regional variations observed by Sziki *et al* (2003). Their analysis, employing micro-PIXE, examined pottery samples with white encrustations from diverse regions in Hungary. They found that samples from Vors-Máriaasszonysziget were predominantly composed of bone, while those from Balatonfuző likely consisted of limestone, and those from Baradla Cave (Aggtelek) were suggested to be made of kaolinite. They demonstrated that variations in white-infilled decorations can occur in regional or temporal contexts and suggested that the elemental composition of the decoration may serve as a potential marker of the pottery's origin.

The following table (**Table 8**) summarises the information from the studies reviewed in detail in this section, presenting the source, sample locations, analytical methods used and identified minerals.

Source	Sample Locations	Analytical Methods	Minerals
Scarcella *et al* (2011)	Capo Alfiere, Perriere Sottano (Italy)	Petrography, SEM-EDS, XRD	Micritic calcitic paste
Scarcella (2011)	Capo Alfiere, Perriere Sottano, Skorba (Malta)	Petrography, SEM-EDS, XRD	Calcium carbonate-clay mixtures
Giustetto *et al* (2013)	Castello di Annone (Piedmont, Italy)	FTIR, Raman spectroscopy, XRPD	Talc, Bone White (calcined hydroxyapatite), mixture of talc and hydroxy-/carbonate-hydroxyapatite, impurities (quartz, micas, feldspars).
Perišić *et al* (2016)	Pločnik (Serbia)	XRPD, FTIR, XRF	Calcium carbonate, crushed bone.
Pirovska (2021)	Western, Central, Eastern Bulgaria	XRD	Calcite, gypsum, talc, barite, dolomite in varying compositions
Gâță and Mateescu (1987)	Vădastra and Crușovu (Romania)	IR, XRD	Quartz, calcite, illite, kaolinite, feldspars, iron oxides, manganese, aluminium
Niculescu (2003)	Boian sites, Călărași County (Romania)	XRD	Calcium carbonate, quartz, gismondine, diopside
Dragoman *et al* (2019)	Spanțov phase sites (Romania)	PIXE, XRD	Calcium compounds, quartz, aluminium-silicates
Opriș et al (2019, 2022)	Sultana, Vidra, Vlădiceasca (Romania)	PIXE, EDX, XRD, FTIR, SEM, EPR	Calcite, bone (hydroxyapatite), quartz, silica-rich sediments

Table 8: Summary of reviewed scientific studies on white-infilled decorations including the sources, sample locations, analytical methods and identified minerals

Chapter 3: Methodology

This chapter outlines the methodology employed in the scientific analysis of the selected pottery sherds.

3.1 Sample Selection

The selected sample set for this study comprises five Għar Dalam phase pottery sherds, all of which display white-infilled decorations. Three fragments were discovered at Santa Verna in 2015, while the other two were found at Skorba in 2016. It is worth noting that all of these fragments were excavated during the FRAGSUS research project (https://www.um.edu.mt/arts/classics-archaeo/ourresearch/fragsus/). The use of recently excavated material was essential for this study, as tampering of decorations on prehistoric pottery sherds by adding white material to enhance the visibility of decorations in photographs was practiced until at least the 1930s, as demonstrated by Luigi Ugolini's work (Pessina and Vella 2021: 215). The use of recently excavated material effectively mitigated this concern.

The scientific analyses (OM, SEM-EDS and micro-XRD) were carried out at the Heritage Malta's Diagnostic Science Laboratories (DSL). Generic information for each sherd, including the site it originated from, the sherd type, type of decoration, identification number, trench identifier and stratigraphic unit can be found in **Table 9**. A detailed description of the sherds can be found in the **Results** section.

Sample Number	Site	Sherd Type	Decoration	Identification Number	Trench	Stratigraphic Unit
1	Skorba	Body	Chevrons	G1002	2016 Trench	23
2	Santa Verna	Body	Net	G1004	D	63
3	Santa Verna	Body/Shoulder	Cardial	G1005	D	119
4	Skorba	Body/Shoulder	Line of 'C' Pattern	G1021	2016 Trench	26
5	Santa Verna	Body	Band of Diagonal Lines	G1030	D	63

Table 9: Sample List (data for table taken from Richard-Trémeau and Betts Unpublished)

3.2 Macroscopy and Section Photography

All the sherds analysed in this research project were previously subjected to macroscopic photography and fabric analysis as part of the MaltaPot project. The MaltaPot team also took microphotographs of the ground edges of the sherds. Each macroscopic photograph is accompanied by a scale, providing dimensions and colour scale for reference. An important role of macroscopic photography is to digitally capture sherds before commencing any analyses, which might impact the artefacts.

3.3 Optical Microscopy

For this study, optical microscopy was conducted using an Olympus BX50 system microscope (**Figure 31**) to capture low-magnification images of the sherds. Photographs were taken at both 4x and 10x magnifications, with a primary focus on examining the white paste, including its texture, inclusions and colour. Another purpose for optical microscopy was to document the microstructure of the white infill under low-power magnification (4-10x).

Figure 31: The Olympus BX50 system microscope used in this study

The captured photos of each sherd were stitched together using DeltaPix InSight software, version 6.0.0, employing image stacking to enhance the depth of focus, resulting in the final composite of the white infill with all the white paste and surrounding ceramic matrix in focus. This process was carried out for images taken at both 4x and 10x magnification.

It should be noted that the images obtained at 10x magnification exhibited a slight reduction in clarity compared to those captured at 4x. Due to the limited depth of field, the optical microscope was unable to achieve consistent focus across the entire surface area of the white paste under 10x magnification.

The observed inclusions within the white infills were compared with Quinn's (2013) classification system, focusing on shape (**Figure 32**) and visual distribution percentage (**Figure 33**). The size of the inclusions was also noted.
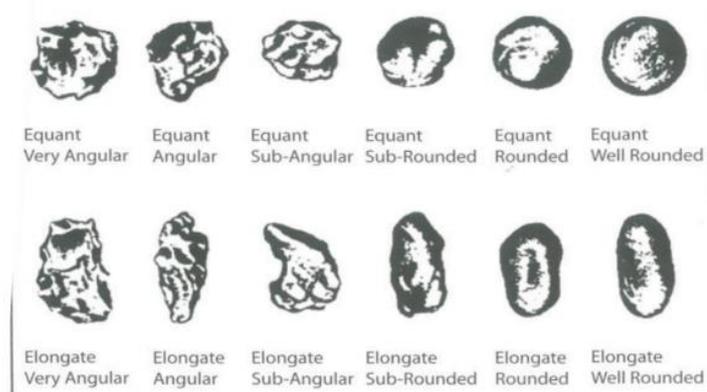
Figure 32: Inclusion Shape Classification System (based on Quinn 2013: 84)
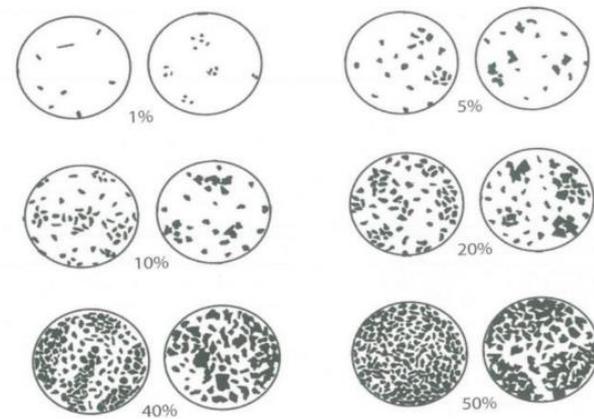
Figure 33: Visual comparison chart for the percentage of microscopic inclusions (Quinn 2013: 82)

3.4 SEM-EDS

The Scanning Electron Microscope (SEM) can serve as a valuable tool for generating highly detailed micrographs of pottery surfaces, capturing features at both microscopic and potentially also macromolecular scales (Froh 2004: 159). When combined with EDS, SEM not only provides qualitative insights but also offers quantitative data regarding the elemental composition of the sample (Froh 2004: 159).

This study utilised a Zeiss EVO MA15 VP-SEM coupled with an Oxford Instruments X-MAX 50 mm^2 EDS detector (**Figure 34**). This model, capable of operating at variable pressures (VP), exhibits enhanced tolerance for non-conductive samples like ceramics, allowing for greater flexibility in sample preparation. Due to this, no coating of the samples was necessary. Rather, the sherds were directly placed on the SEM-EDS holder (**Figure 35**).

After locating the white-infilled decorations on each pottery sherd using the SEM, backscattered electron images were recorded. Most of the micrographs were taken at magnifications ranging from 64x to 282x, with one exception at 1.13 Kx. The imaging process and data storage were managed using the Smart SEM software version 6.01.

Figure 34: SEM-EDS used in this research project
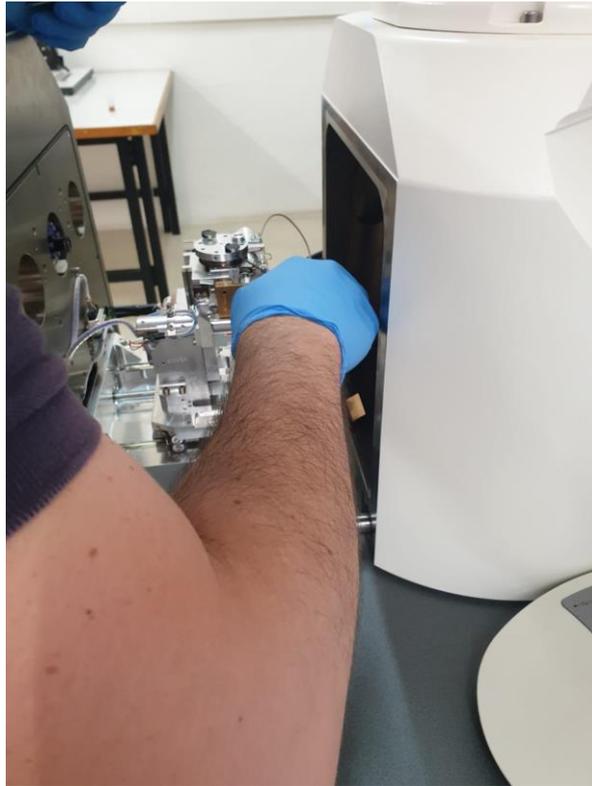
Figure 35: SEM-EDS sample holder

After capturing microphotographs, EDS analysis was performed to identify the elemental composition of the white infills. Initially, a three-pronged trial approach was employed on sherd G1004 to explore the composition. Firstly, point analysis was utilised to gain a quick assessment of elements present in specific locations. Subsequently, a line scan (run no. 2) was conducted across a designated area to obtain a more comprehensive understanding of elemental variations within that region. Following this preliminary assessment, detailed elemental maps were recorded. SEM-EDS mapping captures both the elemental composition and the spatial distribution of elements across the entire area of interest, facilitating a detailed examination of both the white infill and the underlying ceramic body. This was done to ensure efficient contrast between elements originating from the ceramic and infill. The chemical analysis of the pottery matrix itself also offers insights into variations in the materials used, potentially indicating the employment of different clay sources.

The average running time for SEM-EDS mapping was two hours. The EDS data results were presented using two distinct approaches. Firstly, colour-coded elemental maps were generated using the EDS Aztec software, version 3.3, where specific colours indicated the presence and concentration of elements within the sample. Secondly, an alternative presentation employed black (dark) and white (light) elemental maps, where white regions

indicated the detection and localisation of elements on separate maps. These dual approaches visually represented the spatial distribution of elements, with a preference for the clarity of the black and white elemental maps.

The investigation of the white infill was conducted with a focus on a specific area for each sherd, representing a potential limitation for this study. It is assumed that the white infill is homogenous, given its crushed and applied nature. Consequently, the decision to analyse only one area of the white infill was made under the assumption that it would provide representative insights into the overall composition. While this approach allowed for a detailed examination of the selected portion, it is important to acknowledge that variations may exist in different areas of the infill. To address this concern, SEM-EDS was used on two different white infill areas from one sherd (G1004).

3.5 Micro-XRD

Micro-XRD analysis followed SEM-EDS analysis to get a better understanding of the minerals present within the white infills. Sample preparation for micro-XRD analysis involved removing white infill from each sherd using a metal scalpel (**Figure 36**). In preparation for micro-XRD analysis, it was imperative to isolate the white infill from the pottery sherds to prevent any potential interference from the fired clay body. This precautionary measure was necessitated by the rotational movements of the micro-XRD tube and detector, during change in diffraction angle, which could inadvertently include the ceramic body in the analysis alongside the desired white infill.

Figure 36: Scraping white infill of the sherd using a metal needle

A few milligrams of white infill were scraped off and carefully deposited in a recess on a disc-shaped amorphous silicon zero background sample holder (**Figure 37**).

Figure 37: Scraped white infill on the micro-XRD sample holder

Moreover, care was exercised during the extraction of the white infill to ensure minimal contact with the surrounding ceramic matrix. Any inclusion of fired clay or temper for the sherds during this process could lead to the detection of their minerals, thus compromising the integrity of the mineralogical data obtained for the infill. By avoiding any scraping or contamination from the ceramic body, the purity of the white infill samples was maintained to obtain accurate mineralogical information.

The white infill extracted from pottery sherds was subjected to XRD analysis using a Malvern Panalytical Empyrean µXRD instrument with a copper (Cu) anode. The XRD examination commenced at a starting position of 5° and concluded at an end position of 89°, with each step scan set at 0.0130° with a duration of 1887 seconds per step. The data collection was performed continuously in Theta/Theta goniometer configuration, with the incident X-ray beam characterised by specific parameters including a goniometer radius (240 mm) and distance from the focus to the divergence slit (200.8 mm). The analysis utilised K-α1 radiation with a wavelength of 1.54060 Å.

Data Collector version 7.2a was used to collect the data, while HighScore Plus software, version 5.0 (5.0.0.28712), was used to analyse it. Searches were conducted based on chemical formulas, elements or minerals, and the identified compounds were compared with the unknown peaks. Reference data from the Crystallography Open Database (COD) aided in the interpretation of the unknown diffraction patterns. Following this, Rietveld refinement was applied to each diffraction pattern in order to fit each phase and establish a quantity based on the signal recorded.

In line with SEM-EDS methodology, micro-XRD analysis likewise concentrated on a limited quantity of white infill, presumed to be representative of the entire sample. Similarly, this approach presents a potential limitation, as variations may exist within the white infill, as previously discussed in the context of SEM-EDS analysis. To address this, Micro-XRD analysis was conducted on two white infill samples extracted from different areas of pottery sherd G1004 to determine if there were any mineralogical variations between them.

Chapter 4: Results

Macroscopic analysis of the Għar Dalam phase sherds revealed that the white infill consisted of a white/cream, occasionally accompanied by a yellow material. Additionally, crystals were observed within the infill, and in one instance (G1030), microfossils (*foraminifera*) were noted. Variations were noted in the quantity of white infill present in the sherds, leading to the identification of three distinct types:

  i.   Minimal amount of white infill (G1002 and G1005). These sherds exhibited sparse occurrences of white infill in the incisions, manifesting as small patches or specks dispersed sporadically.

 ii.   Moderate amount of white infill (G1030). In this case, the white infill was more noticeable with some patches in the sherd's incisions.

iii.   Substantial amount of white infill (G1004 and G1021). In these cases, the white infill occupied a substantial portion of the sherd's incisions/impressions. Approximately half of the incisions in sherd G1004 were filled with white infill, while all incisions in sherd G1021 were entirely infilled with the white material.

The results will be presented in the following order: first, the macroscopic and section photos. Next, the optical microscope images, followed by the SEM-EDS data and finally, the Micro-XRD data. This organisation is intended to facilitate ease of comparison among the different types of analyses. Additionally, this structured approach allows for a logical progression from macroscopic to microscopic analyses, ensuring a comprehensive examination of the white infills.

4.1 Macroscopy and Section Photography

Detailed visual and descriptive analyses of the sherds utilised in this research project will be presented in this section, providing insights into their macroscopic features and characteristics. These data, previously collected from the MaltaPot team, includes macroscopic photos depicting the exterior and interior of each sherd, with the exception of G1021, for which only the exterior image is available. These photographs will be accompanied by ground edge images for each sample. Each sherd is described in detail, including the following characteristics:

- Sherd type
- Typology
- Macroscopic ware classification
- Wall thickness
- Surface treatment
- Surface interior colour
- Surface exterior colour
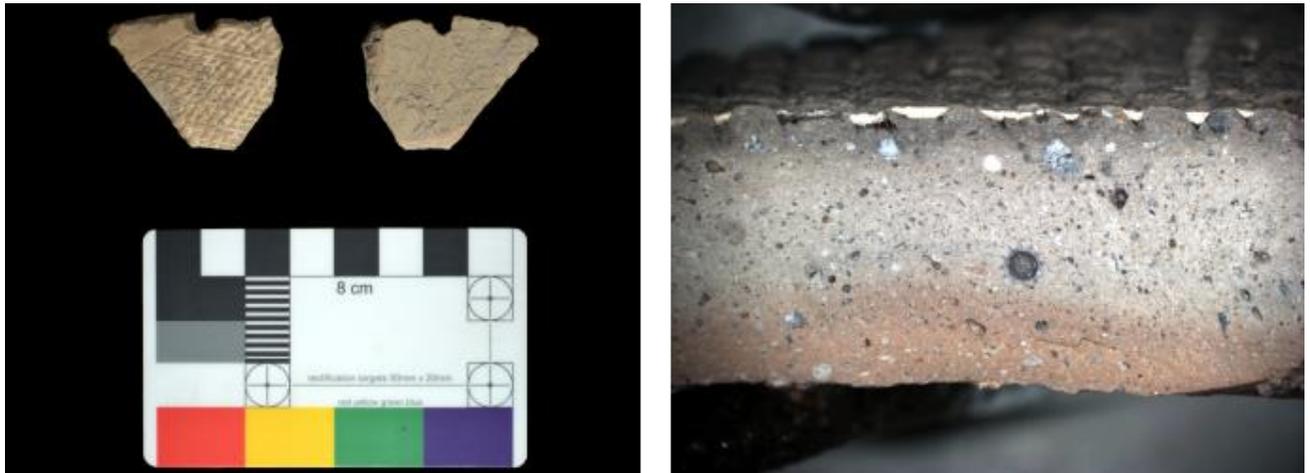- Core colour
- Margins colour
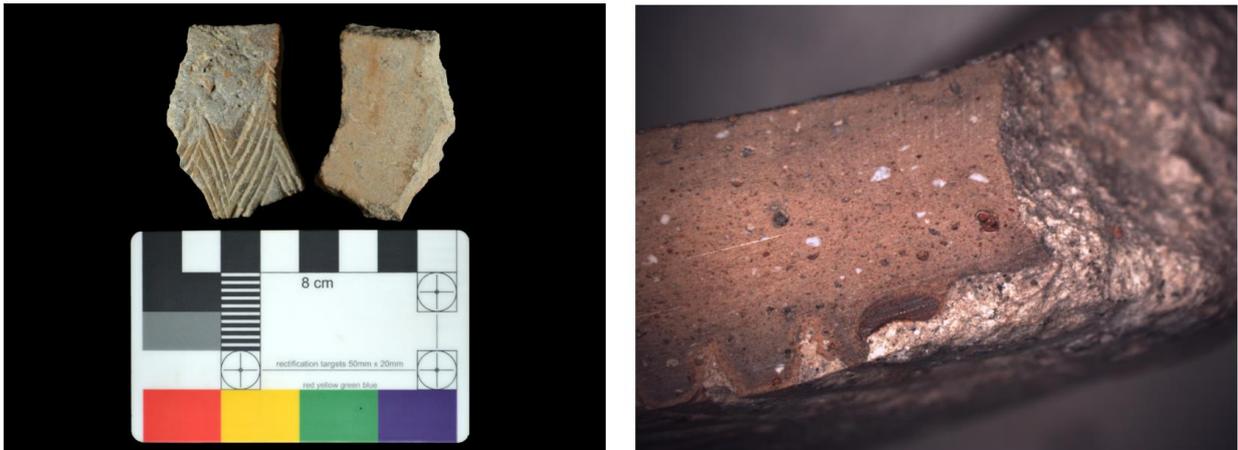
Figure 38: Macroscopic photos of G1002 showing the exterior and interior of the sherd (left) and a microphotograph showing the ground edge of G1002 (right) (Richard-Trémeau *et al* 2023b: 6)

Sherd type	Body sherd
Typology	Evans Għar Dalam type 3 or 4
Macroscopic Ware classification	Fine ware
Wall thickness	7.26 mm
Surface treatment	Decorated with chevron patterns on exterior surface; smoothened interior surface
Surfaces Interior	10YR 7/3 Very pale brown
Surfaces Exterior	10YR 5/1-5/3 gray to brown
Core colour	7.5YR 6/4 light brown
Margins colour	7.5YR 6/1 gray; 7.5YR 6/4 light brown

Table 10: Description of G1002 (Richard-Trémeau *et al* 2023b: 6)

4.1.2 G1004

Figure 39: Macroscopic photos of G1004 showing the exterior and interior of the sherd (left) and a microphotograph showing the ground section of G1002 (right) (Richard-Trémeau *et al* 2023b: 7)

Sherd type	Body sherd
Macroscopic Ware classification	Fine ware
Wall thickness	6.65 mm
Surface treatment	Incised/impressed criss-cross pattern with white paste filling on exterior surface
Surfaces Interior	2.5Y 6/4 light reddish brown
Surfaces Exterior	2.5Y 6/3 light reddish brown
Core colour	10YR 7/2 light grey
Margin colour	10YR 7/2 light grey and 6/4 light yellowish brown

Table 11: Description of G1004 (Richard-Trémeau *et al* 2023b: 7)

4.1.3 G1005

Figure 40: Macroscopic photos of G1005 showing the exterior and interior of the sherd (left) and a microphotograph showing the ground edge of G1005 (right) (Richard-Trémeau *et al* 2023d: 4)

Sherd type	Body sherd
Macroscopic Ware classification	Transitional ware
Wall thickness	8.08 mm
Surface treatment	(Shell?)-impressed decorations with white paste filling
Surfaces Interior	10YR 7/2 light gray
Surfaces Exterior	10YR 6/3 pale brown
Core colour	10YR 3/1 very dark gray
Margin colour	2.5Y 5/1 gray

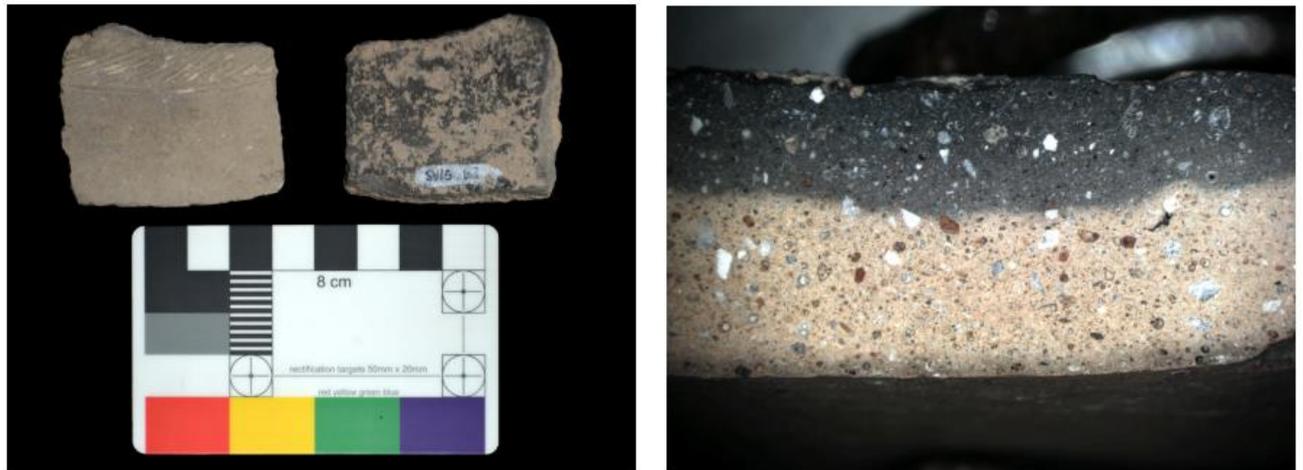**Table 12:** Description of G1005 (Richard-Trémeau *et al* 2023d: 4)

Figure 41: Macroscopic photos of G1021 showing the exterior of the sherd (left) and a microphotograph showing the ground edge of G1021 (right) (Richard-Trémeau *et al* 2023c: 9)

Sherd type	Body/ shoulder
Macroscopic Ware Classification	Fine ware
Wall thickness	6.56 mm
Surface treatment	Band of impressed "C" with white infill
Surface Interior	2.5Y 3/1 very dark grey
Surface Exterior	2.5Y 3/1 very dark grey
Core Colour	2.5Y 5/1
Margin Colour	2.5Y 4/1

Table 13: Description of G1021 (Brogan *et al* Unpublished)

Figure 42: Macroscopic photos of G1030 showing the exterior and interior of the sherd (left) and a microphotograph showing the ground edge of G1030 (right) (Richard-Trémeau *et al* 2023b: 12)

| Sherd Type	Body sherd
Macroscopic Ware classification	Fine, with some white inclusions
Wall thickness	7.93 mm
Surface treatment	Impressed or incised parallel lines. White paste.
Surfaces Interior	10YR 4/1 dark gray
Surfaces Exterior	10YR 6/2 light brownish gray
Margin Colour	10YR 7/3 very pale brown and 10YR 4/1 dark gray

Table 14: Description of G1030 (adapted from Richard-Trémeau *et al* 2023b: 12)
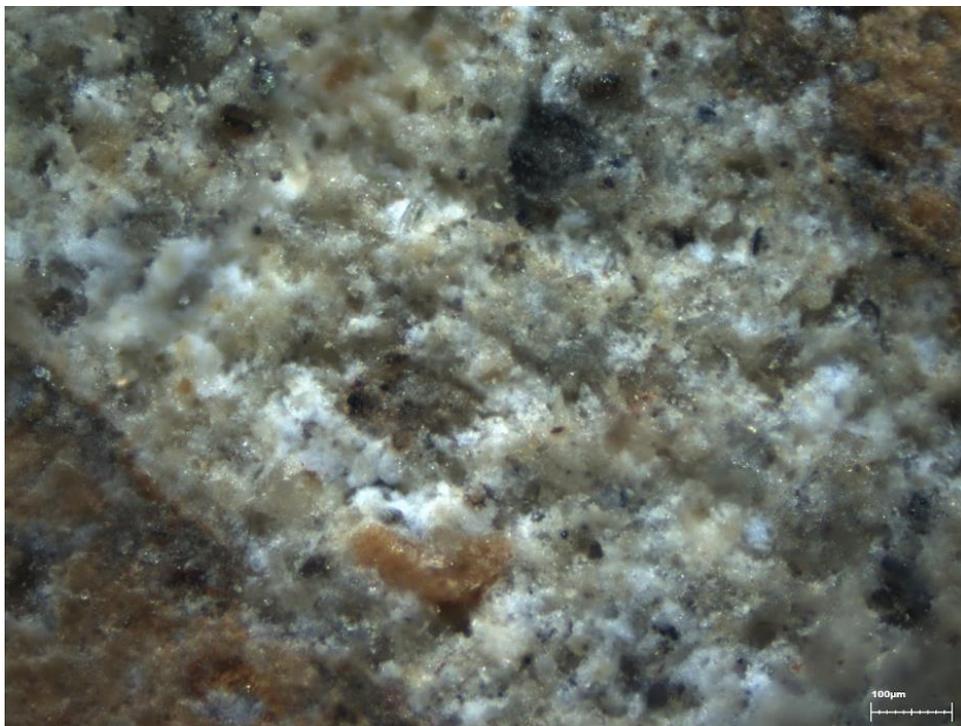
4.2 Optical Microscopy

In this section, the optical microscope results will be presented, including images captured at both 4x and 10x magnification. During analysis, it was observed that different colours were present within the white infill of some samples. Specifically, the white infills of G1004 (**Figures 45-46**) and G1021 (**Figures 49-50**) appeared to consist entirely of a homogeneous white material under microscopic examination. In contrast, the white infills of G1002 (**Figures 43-44**), G1005 (**Figures 47-48**) and G1030 (**Figures 51-52**) exhibited a more heterogeneous composition, displaying a mixture of white and yellowish material. This yellowish component dominated the white infill of G1002. No atomic contrast was recorded between these two materials in backscatter electron imaging, potentially because they share similar densities.

Additionally, different inclusions were observed within the white infills, including small (<20µm) equant rounded, elongate sub-rounded and elongate angular particles (Quinn 2013: 84). These inclusions were noted in all of the white infills and, according to Quinn's (2013: 82) visual examination percentage comparison chart (**Figure 33**), approximately comprised 1-5 % of the white infill material. Transparent angular and rounded inclusions (10-40 µm) were observed within the white infills, albeit less frequently than the inclusions mentioned above. These inclusions constituted approximately <1 % of the total white infill area. Their presence atop the white infill material suggests post-depositional contamination rather than intrinsic composition within the infill matrix. In only one of the white infills (G1030), a *Globigerina sp.* foraminifera (*c.* 280µm diameter) and another foraminifera (*c.* 240µm diameter) with a rounded shape adjacent to the *Globigerina* were observed (Foraminifera.eu n.d). These specimens appear to be part of the white infill material.
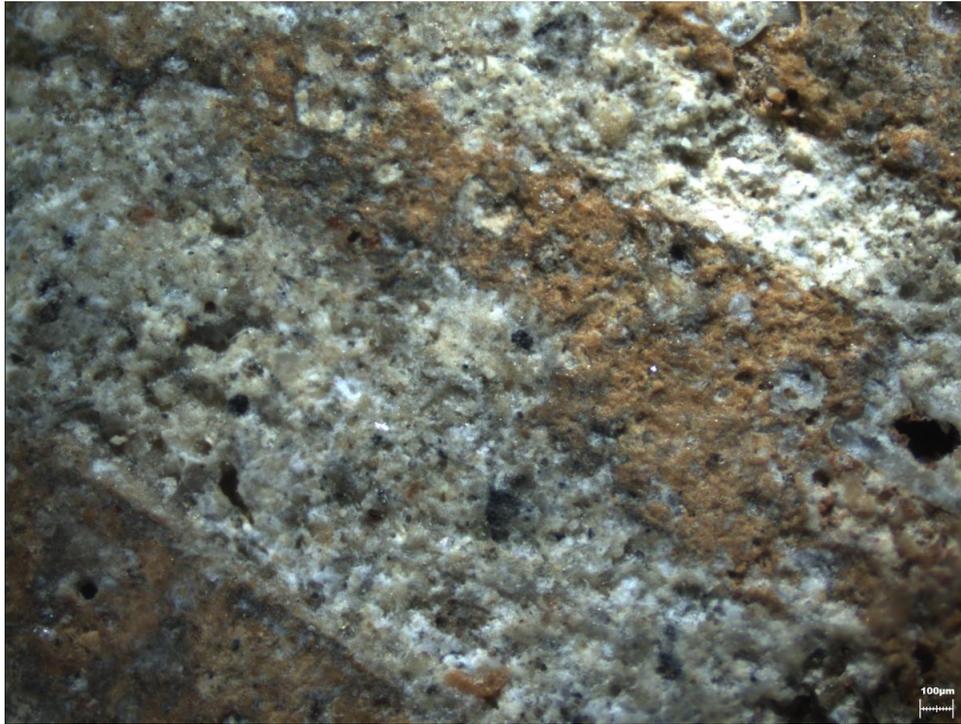
Figure 43: Optical microscope image (4x) showing the white infill and the ceramic body of G1002

Figure 44: Optical microscope image (10x) focusing mainly on the white infill of G1002

Figure 45: Optical microscope image (4x) showing the white infill and the ceramic body of G1004

Figure 46: Optical microscope image (10x) focusing mainly on the white infill of G1004

Figure 47: Optical microscope image (4x) showing the white infill and the ceramic body of G1005

Figure 48: Optical microscope image (10x) focusing mainly on the white infill of G1005

Figure 49: Optical microscope image (4x) showing the white infill and the ceramic body of G1021

Figure 50: Optical microscope image (10x) focusing mainly on the white infill of G1021

Figure 51: Optical microscope image (4x) showing the white infill and the ceramic body of G1030. Two foraminifera specimens are visible in the white infill.
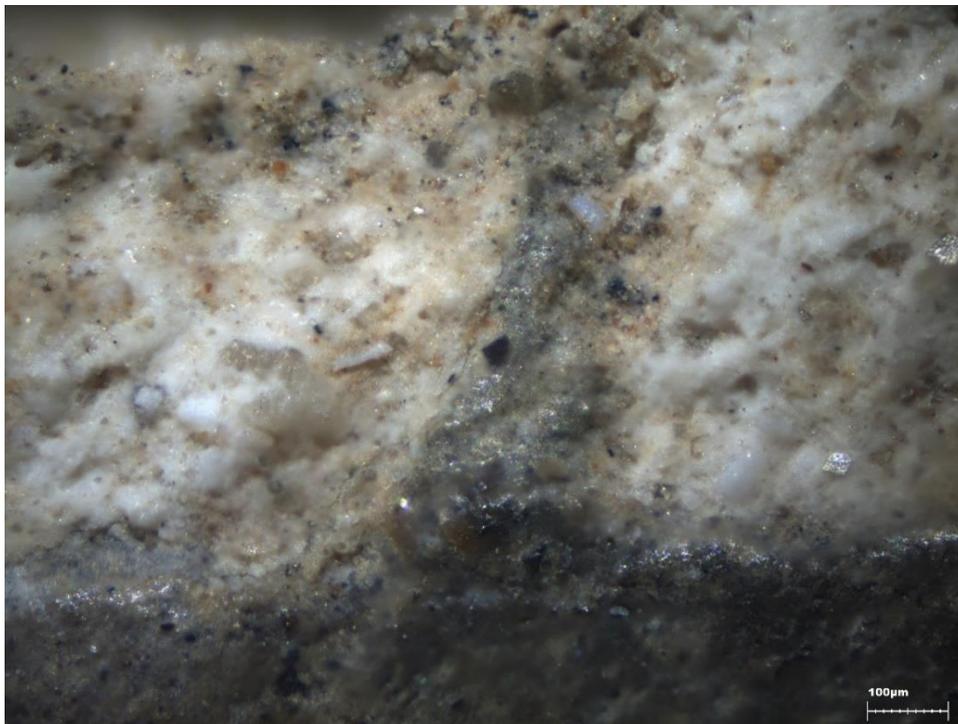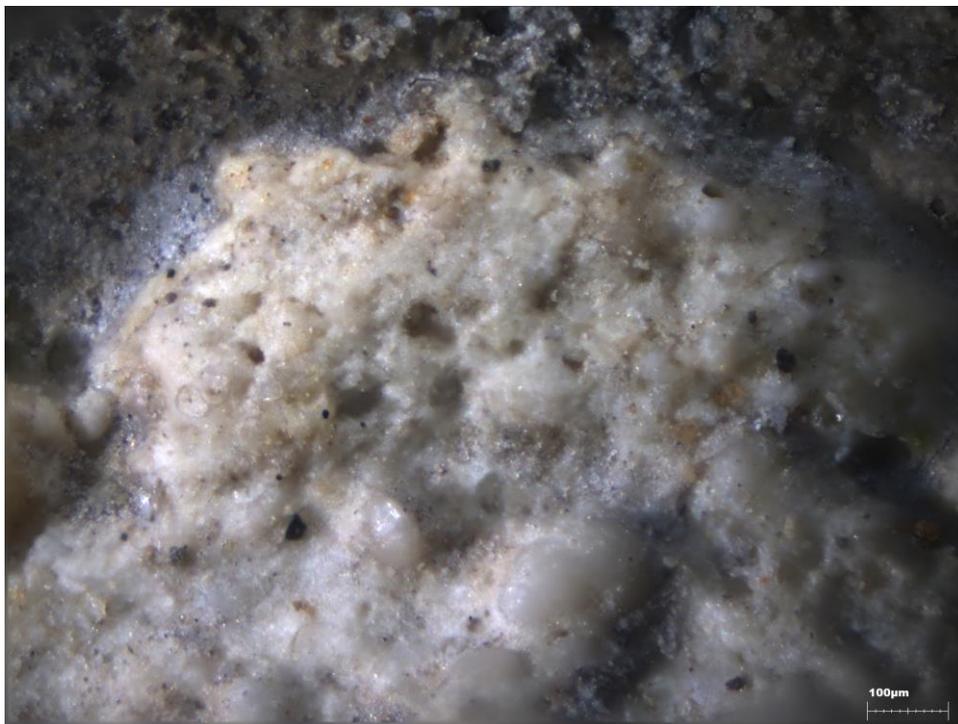

Figure 52: Optical microscope image (10x) focusing mainly on the white infill of G1030

4.3 SEM-EDS

This section presents the SEM-EDS imaging and analysis of the white-infilled decorations. Each subsection details the elemental composition identified by EDS, through mapping, including the specific elements present, their location within the decoration, and their relative concentrations. The backscattered electron images lack clear differentiation between the ceramic body and the white-infilled decorations due to low atomic contrast. This occurs because materials with similar atomic numbers provide less atomic contrast. These images are included in the **Appendix A**.

Calcium was detected using EDS within all the white infills, emerging as the predominant element. Its abundant presence distinctly delineates the locations of the white-infilled decorations (as can be seen in **Figure 54**). Silicon and aluminium were found together within the ceramic bodies of all the sherds analysed. Additionally, significant silicon concentrations were detected in the white infills of G1004 and G1005, while G1030 exhibited a lower relative abundance of silicon. No aluminium was detected in the white-infilled decorations. In samples G1004 (**Figure 54**) and G1005 (**Figure 55**), higher amounts of magnesium were detected, whereas in G1021 (**Figure 56**), it was found in lower amounts. Iron detections were scattered in the ceramic matrix of G1002, G1004, G1005 and G1030, with some intense concentrations. Additionally, some high concentrations were also noted in the white infills of G1002 (**Figure 53**), G1005 and G1030 (**Figure 57**).

Potassium was also detected in low amounts in all of the sherds. Namely, in the matrix of G1002 and G1030, in the infill of G1021 and in both the infill and the matrix G1004 and G1005. The distribution of phosphorus varied among the sherds. In sherds G1002 and G1021, a low concentration of phosphorus was detected within the matrix. Conversely, in sherd G1004, a concentration of phosphorus points, one being significantly more intense than the others, was observed in the white infill, with relatively fewer concentration points present in the ceramic body. In contrast, phosphorus was detected throughout the entire analysed area in G1030, while in sherd G1005, phosphorus emerged as a main element in the white infill. Carbon was detected in relatively low amounts in the ceramic bodies of G1002, G1021 and G1030. In G1004, little carbon was detected in the matrix while a comparatively higher intensity was noted in the white infill. In G1005, higher concentrations of carbon were detected, which were spread throughout the analysed area. However, it is important to clarify the interpretation of these findings. The

presence of carbon detected by EDS signifies concentrations exceeding a specific detection threshold, not necessarily the absence of it elsewhere in the sherd.

Sodium, titanium and sulphur, were consistently detected in very low concentrations. Their presence was very low in the analysed areas of G1002, G1003, G1021 and G1030. G1005 displayed a comparatively higher amount of sodium, present in both the matrix and the white infill. Lastly, singular points of chlorine were observed in the ceramic bodies of G1005, G1021 and G1030 and in the white infill of G1005. The following table (**Table 15**) presents the elements detected in the white infills using EDS.

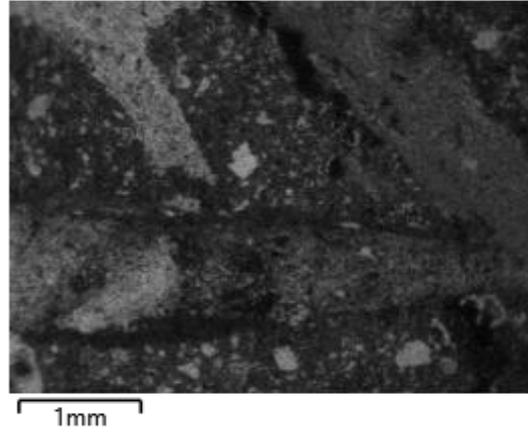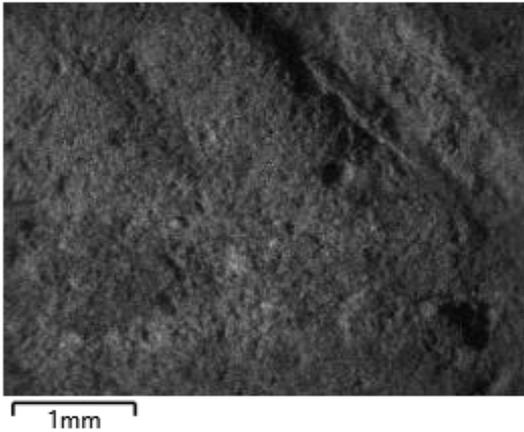| Sample	Ca	Si	Mg	Fe	K	P	C	Na	Cl
G1002	✓			✓					
G1004	✓	✓	✓		✓	✓	✓		
G1005	✓	✓	✓	✓	✓	✓	✓	✓	✓
G1021	✓		✓		✓				
G1030	✓	✓		✓		✓			

Table 15: Elements detected in the white infills through EDS

4.3.1 G1002

Si K series
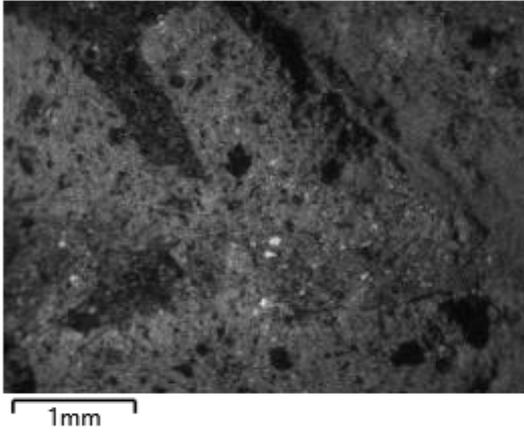
Ca K series

O K series
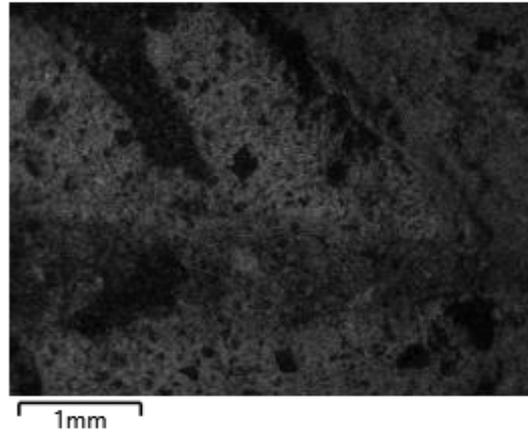
Al K series

K K series
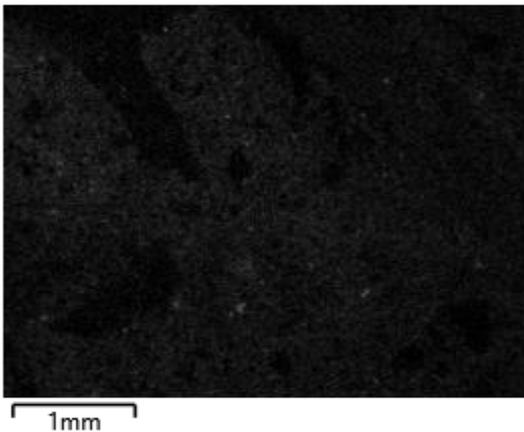
Fe K series
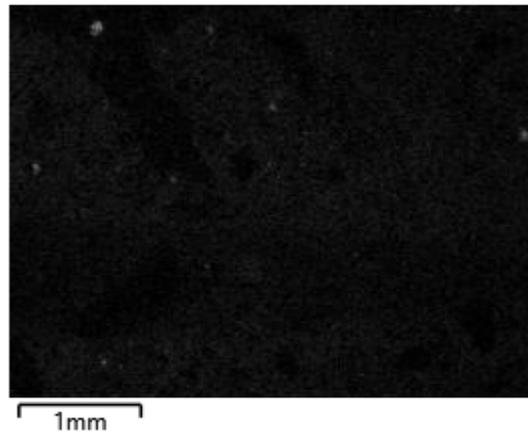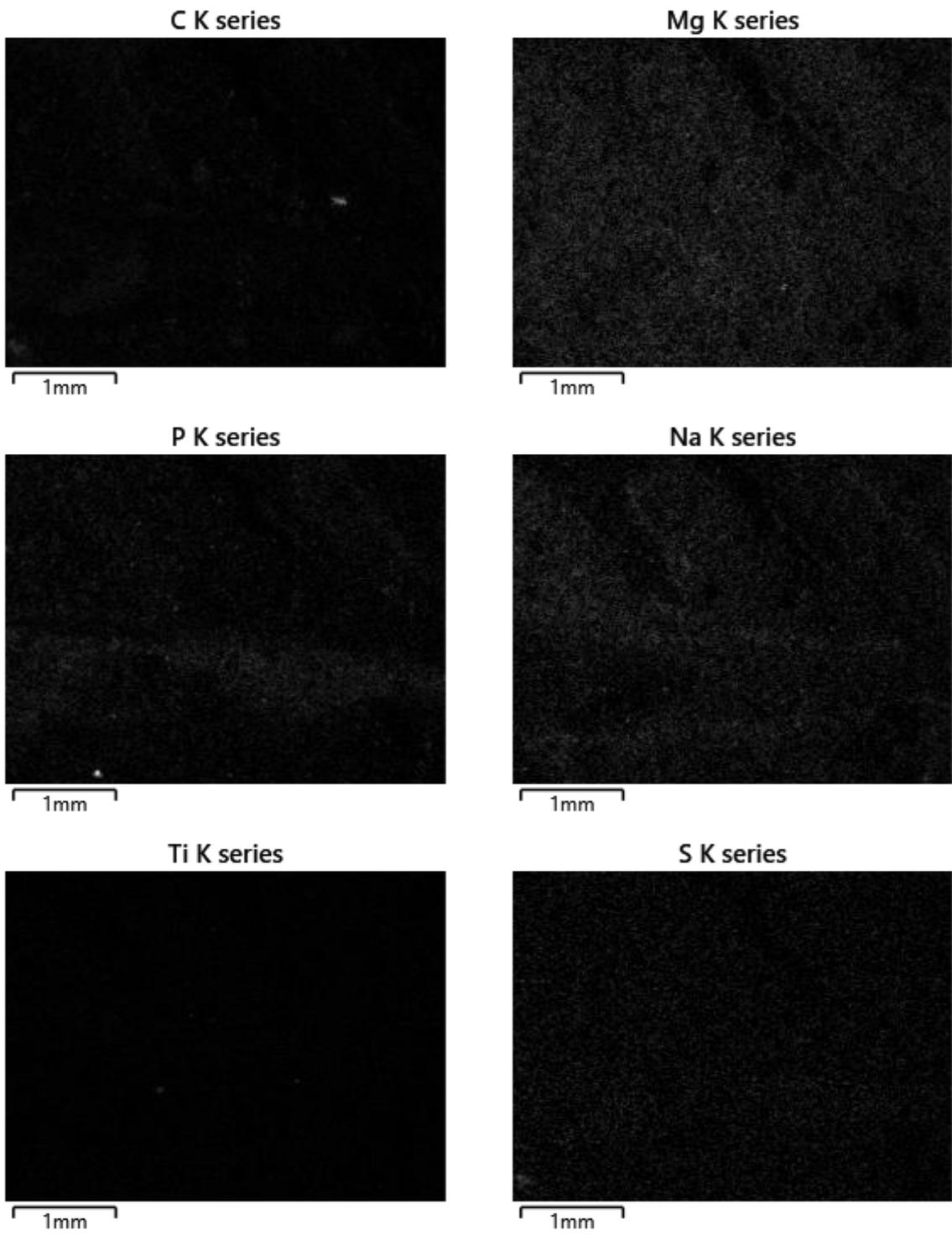
Figure 53: EDS Data of G1002

4.3.2 G1004

Ca K series

1mm

Si K series

1mm

O K series

1mm

Al K series

1mm

K K series

1mm

Fe K series

1mm
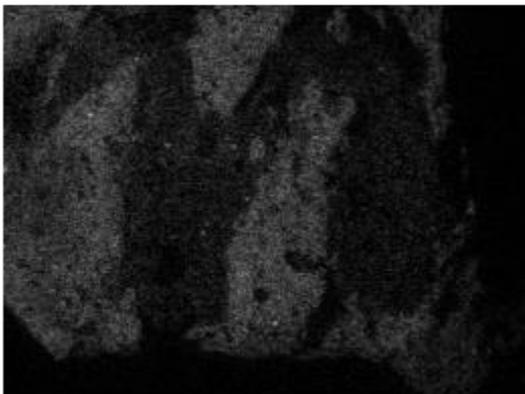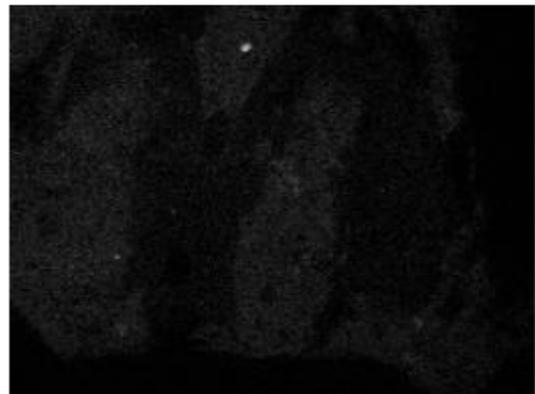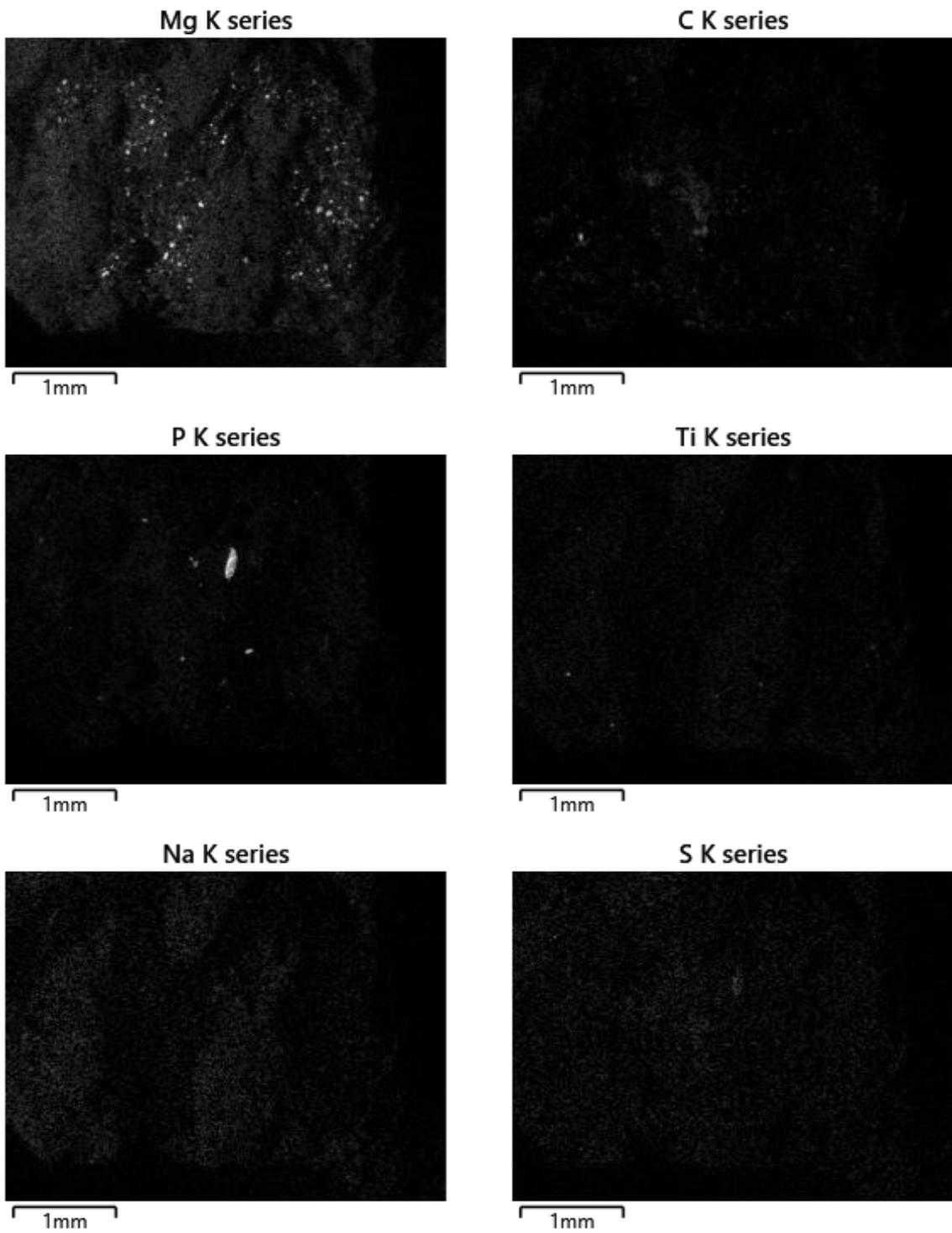
Figure 54: EDS Data of G1004 (analysis no. 1)
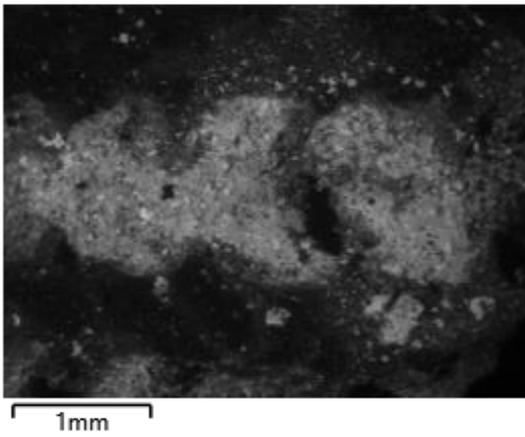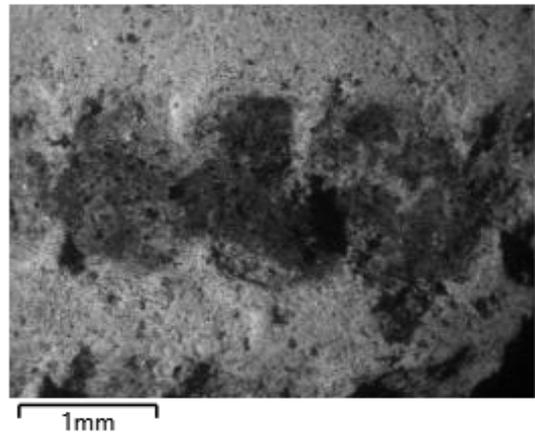
Si K series

1mm

O K series

1mm

Ca K series

1mm

Al K series

1mm

K K series

1mm

P K series

1mm

Fe K series

1mm

Mg K series

1mm

Na K series

1mm

Ti K series

1mm

C K series

1mm

S K series

1mm
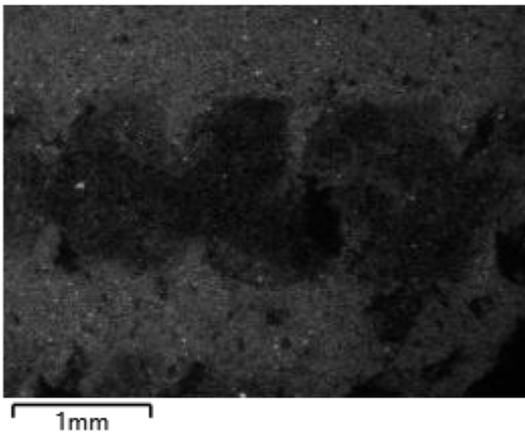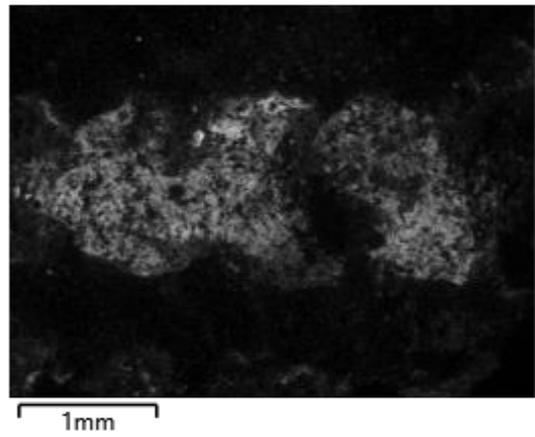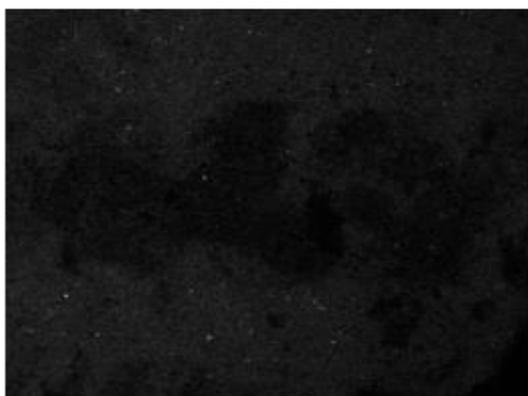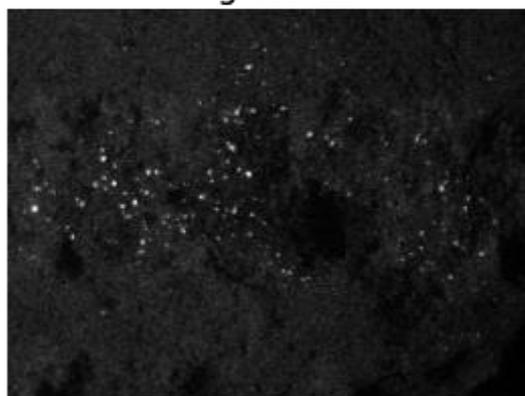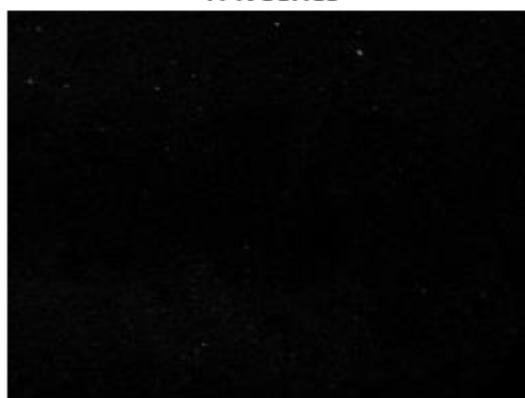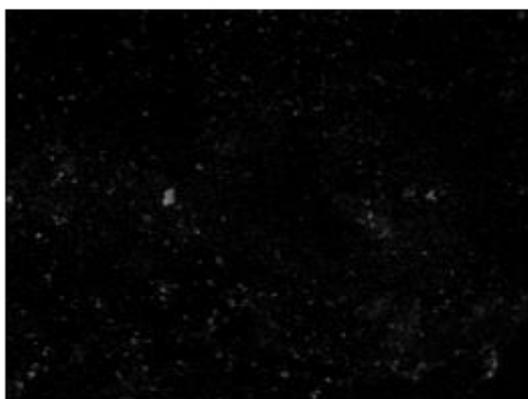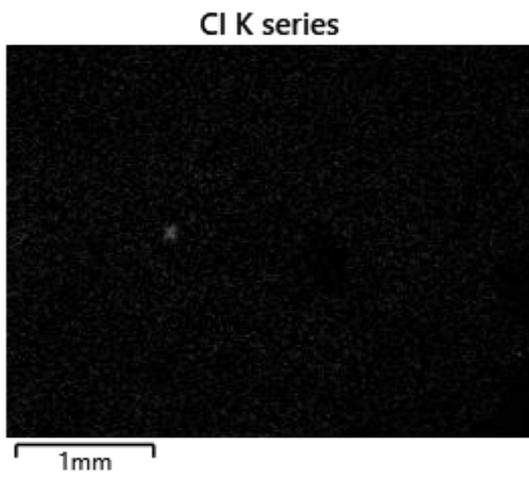
Figure 55: EDS Data of G1005

Ca K series

Si K series

O K series
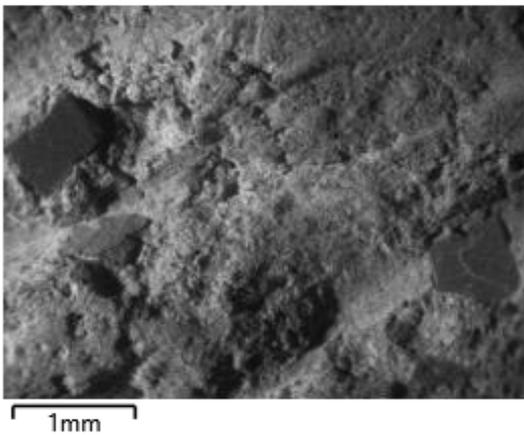
Al K series

C K series

K K series

1mm

1mm

1mm

1mm

1mm

1mm

Figure 56: EDS Data of G1021

4.3.5 G1030

Ca K series

Si K series

1mm

1mm

O K series

Al K series

1mm

1mm

C K series

K K series

1mm

1mm

Fe K series

1mm

Mg K series

1mm

P K series

1mm

Na K series

1mm

Ti K series

1mm

S K series

1mm
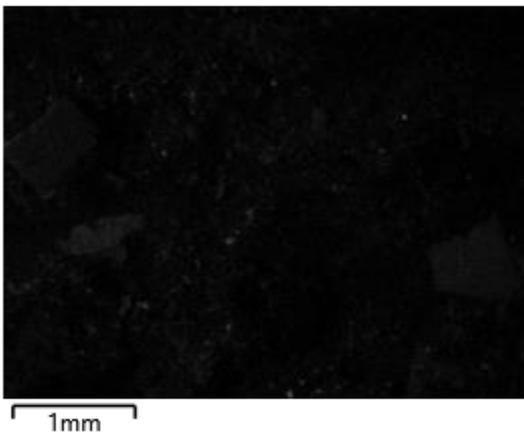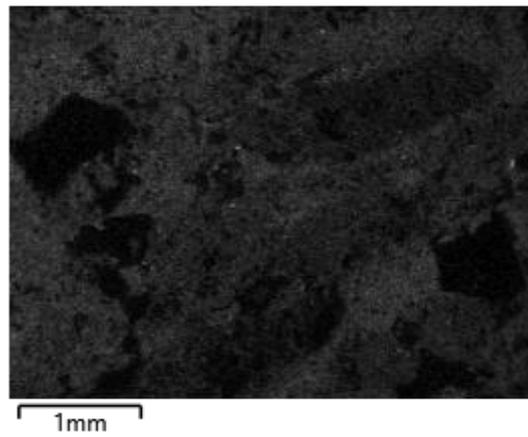
Figure 57: EDS Data of G1030

A distinctly large fragment (**Figure 58**) was detected in the white infill of G1030 during SEM analysis. The fragment measured approximately 830μm in length and exhibited a rectangular shape, with linear features in the middle resembling cracks, and one end engulfed in the ceramic matrix. Its relatively large size led to its serendipitous discovery during SEM-EDS mapping and it was the only one of its type noted during the analysis of the white infills.
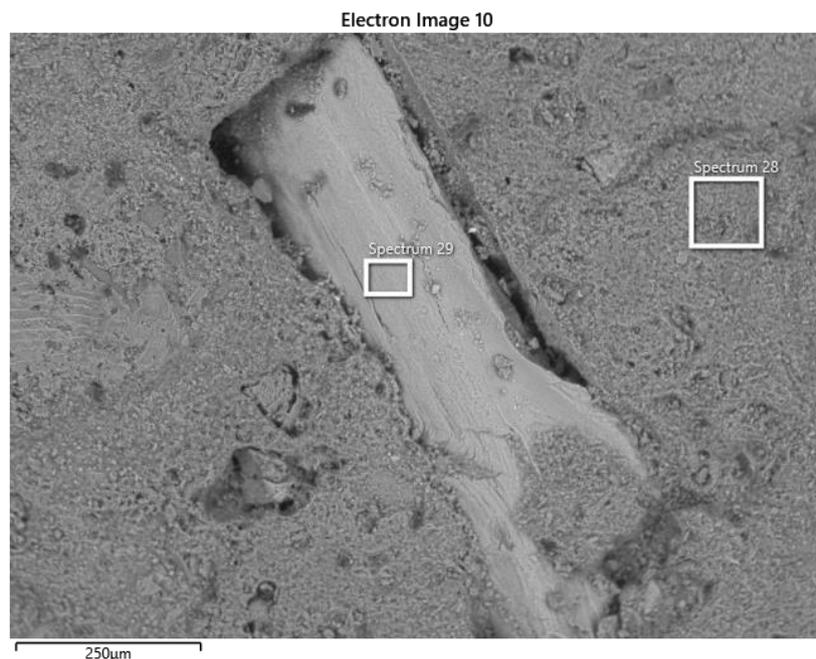
Figure 58: Fragment in the white infill of G1030

This fragment was analysed using the point analysis tool to get a better understanding of its composition (spectrum 29 shown in **Figure 58**). The data showcased relatively high peaks of calcium, phosphorus and oxygen and other smaller peaks corresponding to several other

elements (Ti, C, Ce, Fe, F, Na, Mg, Al, Si, S, Cl and K). From these elements, silicon and aluminium had the largest peaks. Additionally, an area scan of the ceramic body was carried out for this sherd (spectrum 28 shown in **Figure 58**). The data, which is included in **Appendix C (Figure 77)**, shows relatively high peaks of silicon, oxygen, calcium and aluminium and smaller peaks corresponding to other elements (Ti, C, Fe, Na, Mg, P, S and K).
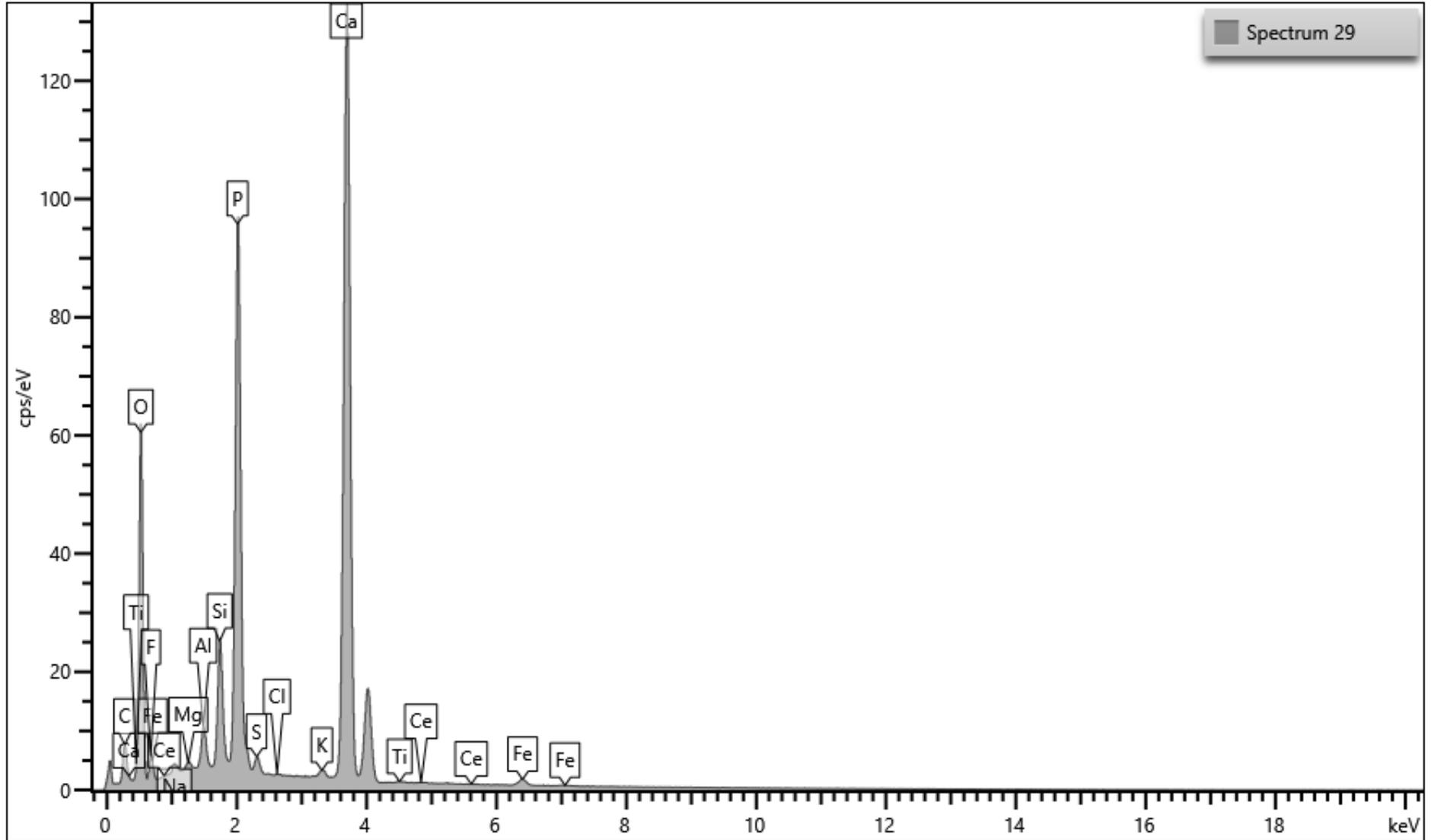
Figure 59: EDS data (spectrum 29) of the fragment in the white infill of G1030

4.4 Micro-XRD

Micro-XRD analysis was conducted on the five samples (G1002, G1004, G1005, G1021 and G1030). Sample G1002 primarily consisted of calcite, with quartz as the second major component. Additionally, aragonite was present in low amounts. Sample G1004 indicated a dominance of calcite, with quartz and dolomite also present but in lower amounts. In sample G1005, calcite was the predominant mineral, while quartz, dolomite and fluorapatite accounted for lower percentages. Sample G1021 was predominantly composed of calcite, with quartz and dolomite present in much lower amounts. Finally, Samples G1030 showed a high calcite content with a low amount of quartz. Refer to **Table 16** for the relative percentages of minerals identified in the white infills. The micro-XRD data for run number 2 of sample G1004, aimed at assessing the homogeneity of the white infills, is provided in the **Appendix B**.

Sample	Calcite	Quartz	Dolomite	Aragonite	Fluorapatite	Rwp Values
G1002	61.1 %	31.0 %	/	7.9 %	/	7.6915
G1004	77.6 %	3.3 %	19.1 %	/	/	10.6636
G1005	51.4 %	6.9 %	19.2 %	/	22.4 %	9.0163
G1021	97.0 %	0.8 %	2.2 %	/	/	8.7748
G1030	92.2 %	7.8 %	/	/	/	8.0296

Table 16: Results of micro-XRD analysis showing the relative percentages of minerals identified in the white infills

4.4.1 G1002
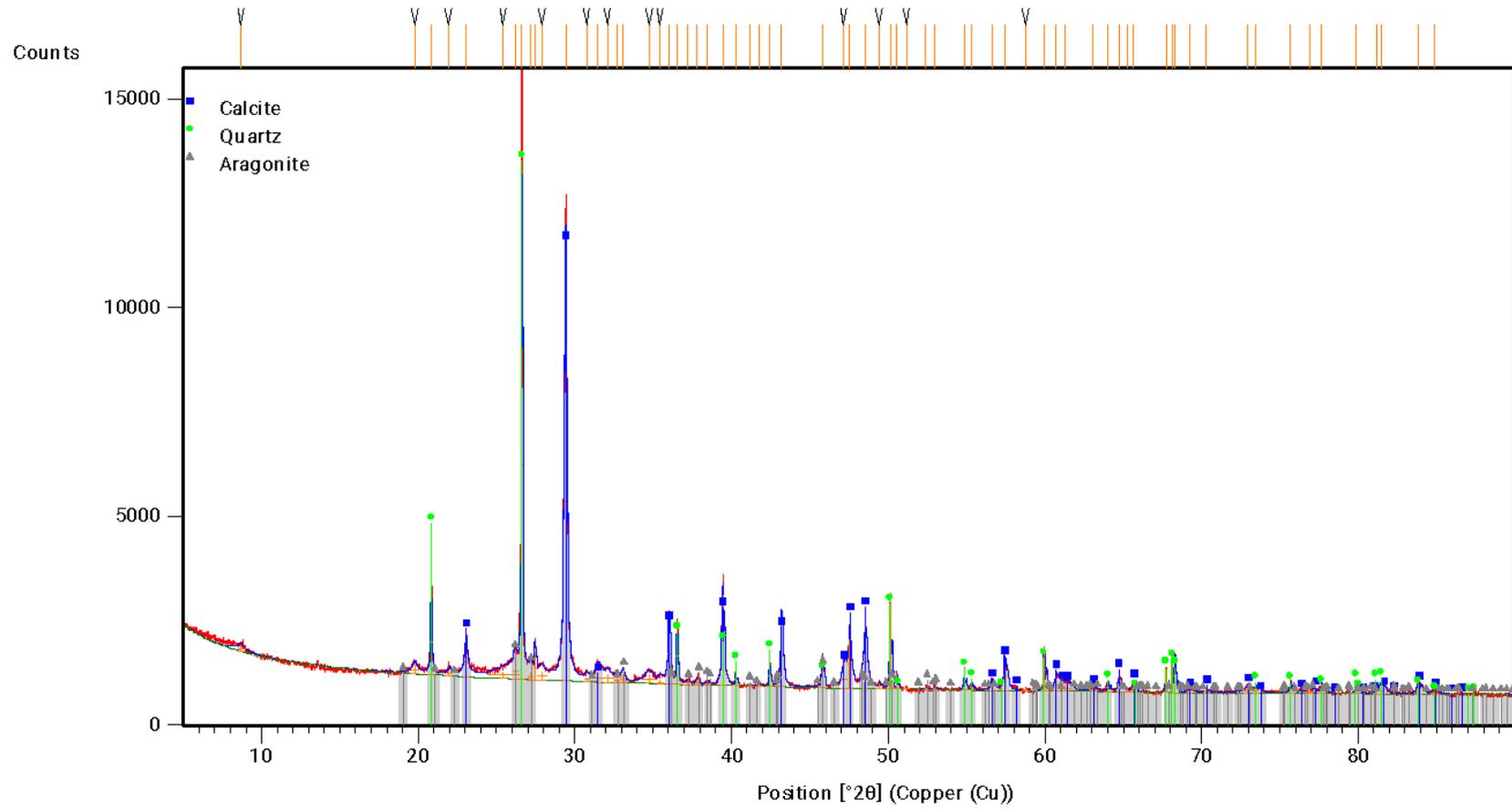
Figure 60: Micro-XRD data of G1002
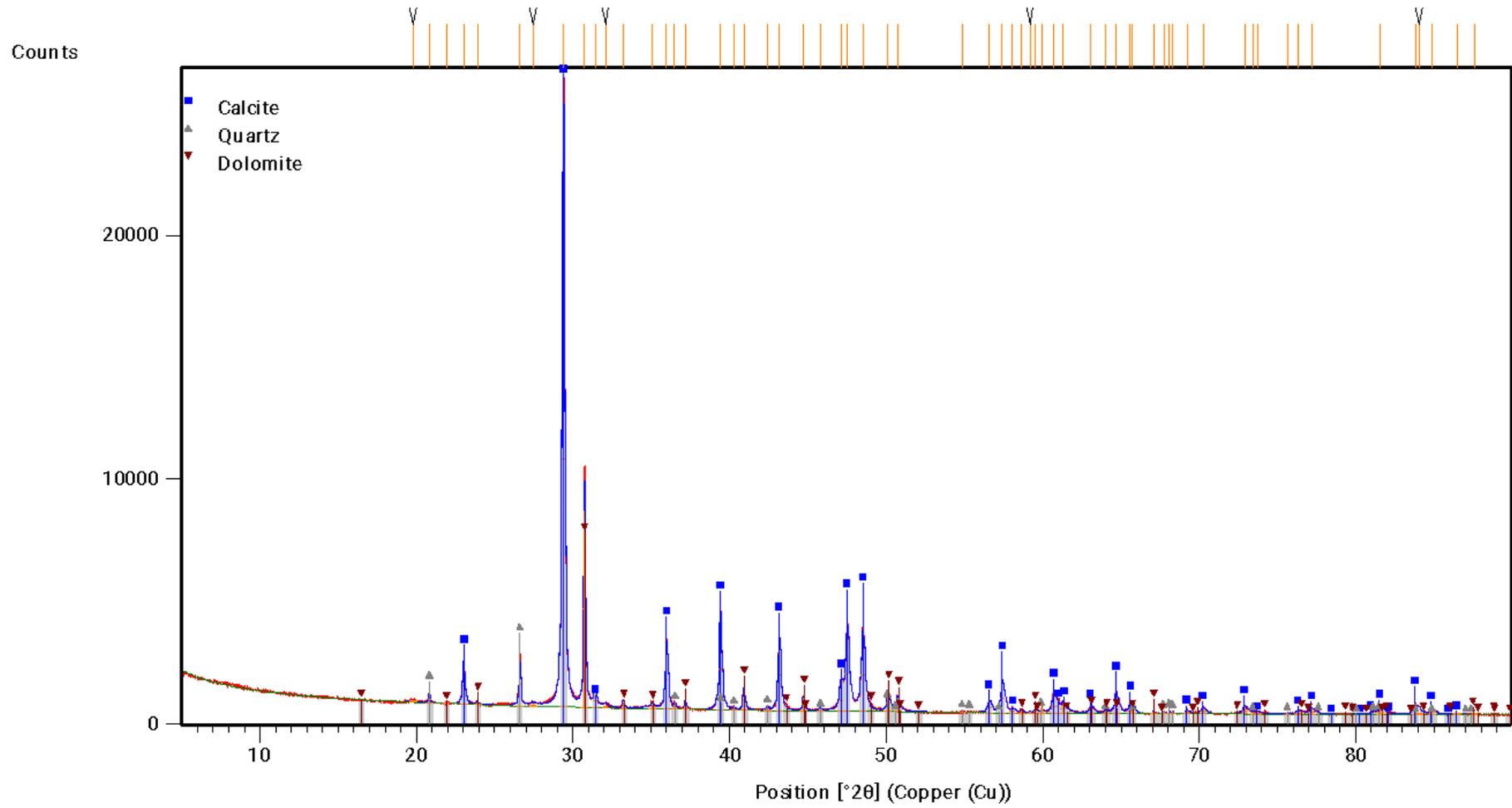
Figure 61: Micro-XRD data of G1004 (analysis no. 1)
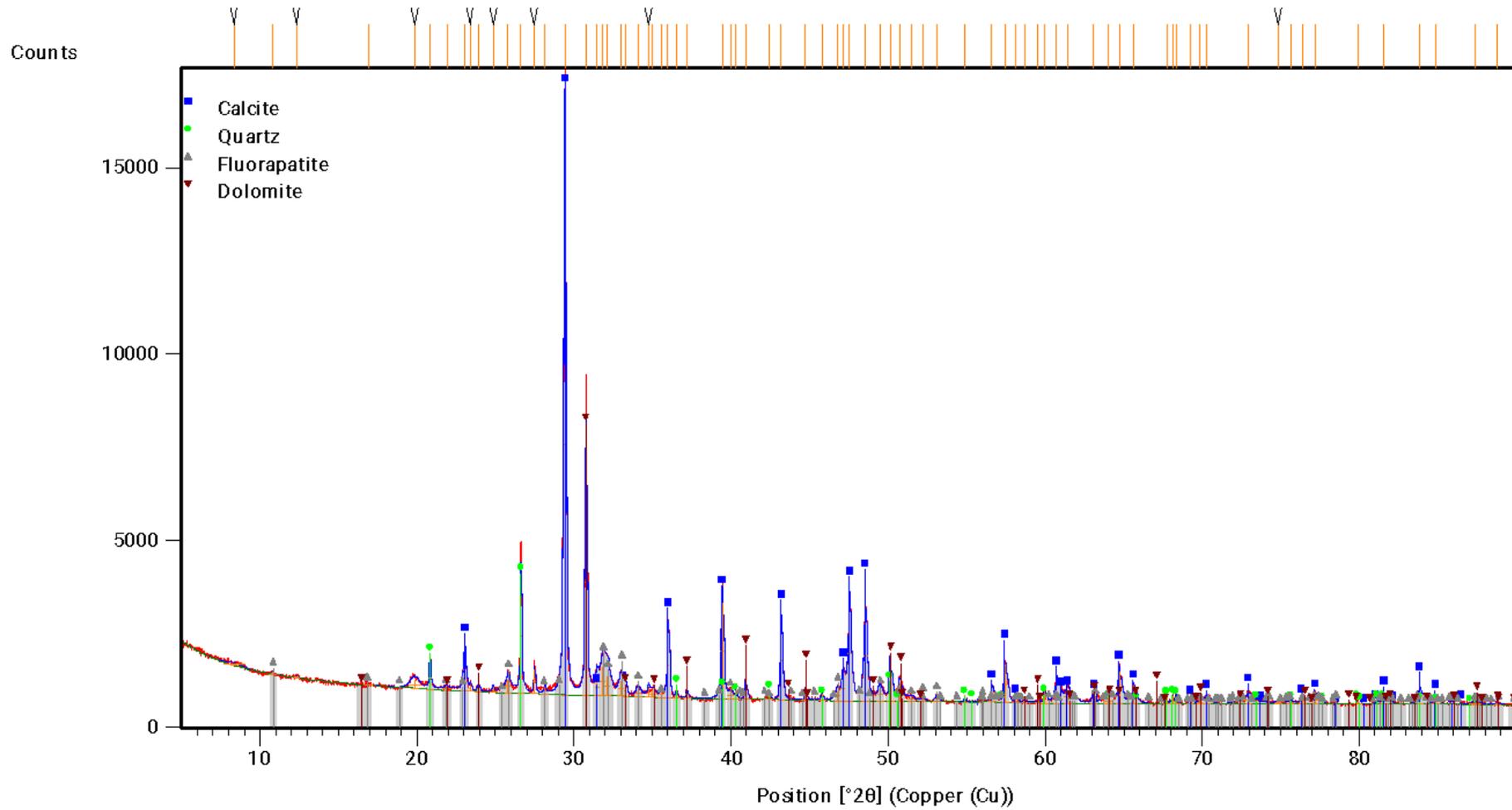
Figure 62: Micro-XRD data of G1005
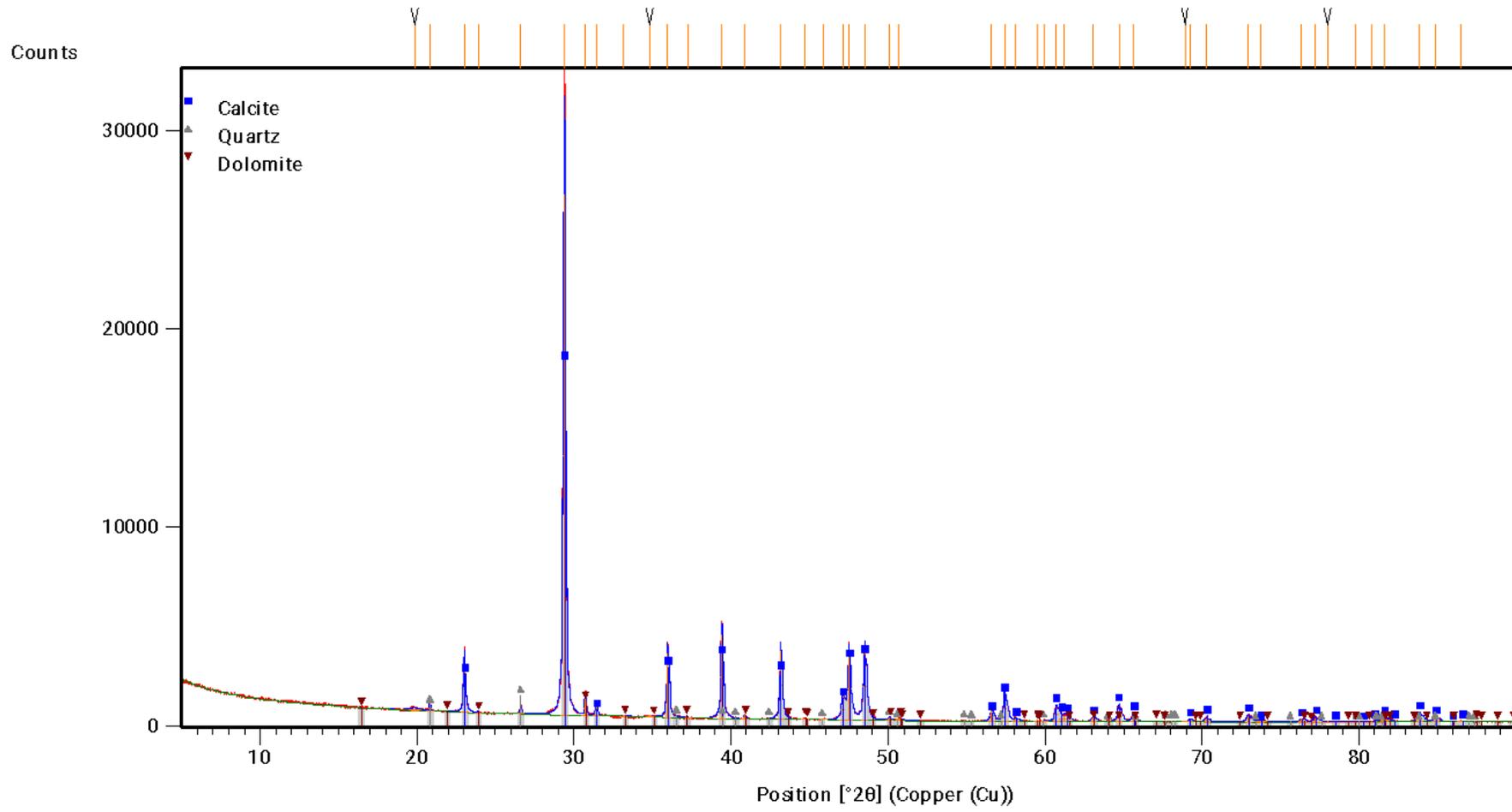
Figure 63: Micro-XRD data of G1021
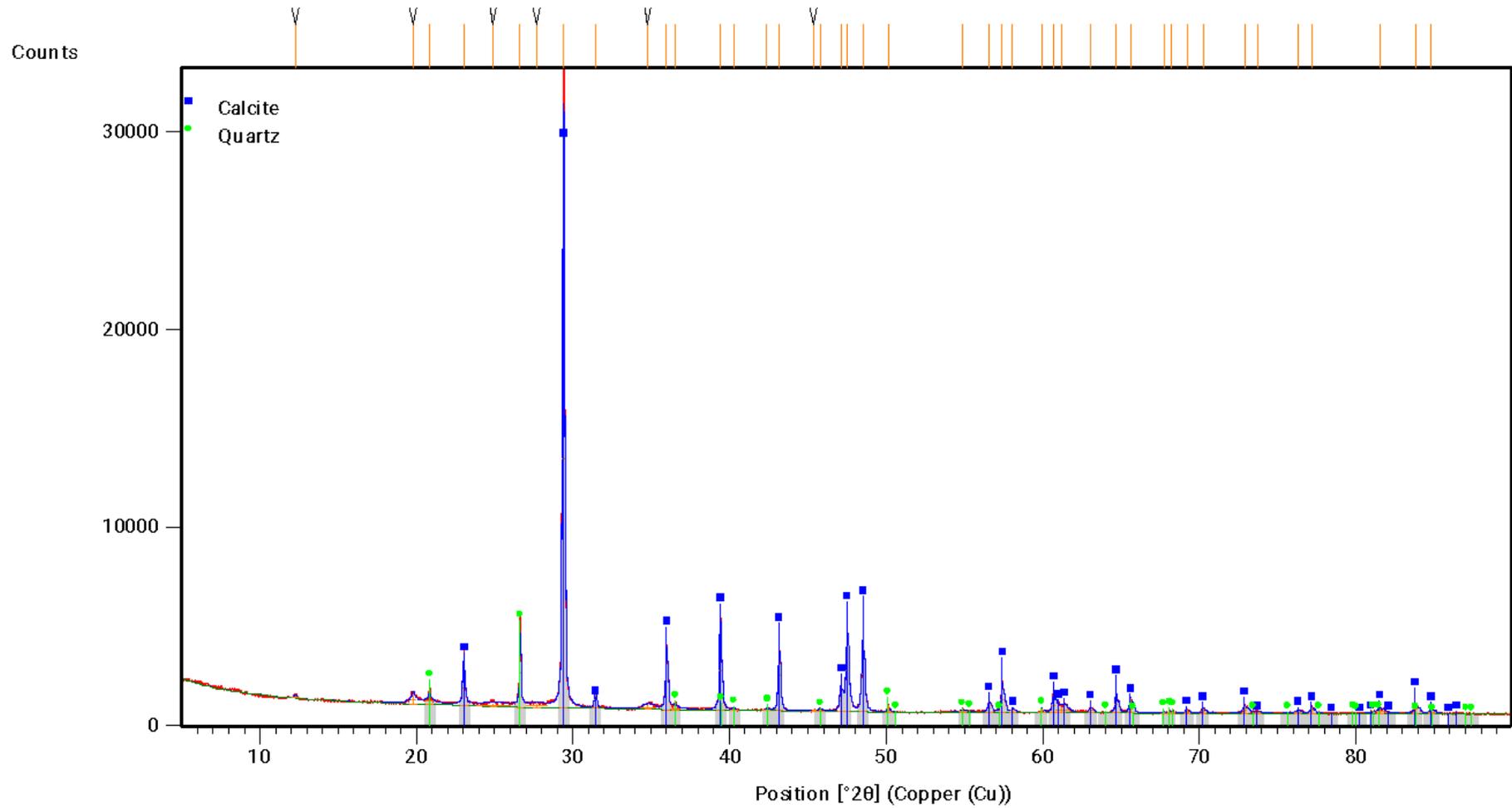
Figure 64: Micro-XRD data of G1030

Chapter 5: Discussion

5.1 The Chemical and Mineralogical Data

The adopted methodology was effective in comprehensively analysing the white-infilled decorations by first acquiring chemical data, through EDS, for both the white infills and the ceramic matrix, followed by micro-XRD to obtain the mineralogical data. EDS analysis revealed that all white infill samples primarily consisted of a high calcium-based component with additional elements present in the white infills. Additionally, EDS mapping showed clear variations between the chemical compositions of the white infills and the ceramic bodies. This non-destructive step was crucial for obtaining an initial understanding of the elements and potential minerals in the white infill, thereby justifying the sampling of the white infill for micro-XRD analysis. Micro-XRD analysis was necessary to determine the mineral composition of the white infill, to get a better understanding of what the white infills were composed of. For instance, while EDS detected high calcium content in the white infills, micro-XRD, indicated that the calcium present in the white infill originated from three different calcium-containing minerals which included calcite, dolomite and aragonite. The presence of magnesium (Mg) detected by EDS could be attributed to the presence of dolomite ($CaMg(CO_3)_2$), as later identified by micro-XRD. Similarly, the relatively high presence of phosphorus detected by EDS in sample G1005 (**Figure 55**) was attributed to the presence of fluorapatite, as confirmed by micro-XRD. The analysis of the white infill in sample G1005 further underscores the necessity of employing both chemical and mineralogical analyses. Despite the absence of fluorine in EDS data, which is a component of fluorapatite, the presence of fluorapatite mineral was identified through micro-XRD. The points made above emphasise the need for complementary analytical techniques to achieve comprehensive understanding.

5.2 Nature of the Minerals in the White Infills

The analysed white infills predominantly consisted of calcite and quartz, among other minerals. This finding contrasts with Trump's (1966: 23) hypothesis, which proposed gypsum as the primary material for the white-infilled decorations in Għar Dalam phase pottery.

Based on the composition, the analysed infills can be categorised into four main divisions: a) Mixtures primarily composed of calcite and quartz as a minor component (G1030); b) mixtures predominantly composed of calcite with minor proportions of quartz and dolomite (G1004 and G1021); c) calcite and quartz dominated infill with small quantities of

aragonite (G1002) and d) white infill primarily composed of calcite but with low levels of quartz, dolomite and fluorapatite (G1005). Despite the archaeological origins of the sherds, retrieved from two locations, Skorba in Malta and Santa Verna in Gozo, all specimens displayed calcite and quartz as the most common components in the white infills. Furthermore, dolomite was detected in sherds from both Skorba (G1021) and Santa Verna (G1004 and G1005). Aragonite was only found in a single sherd from Skorba (G1002), while fluorapatite was solely detected in one sherd from Santa Verna (G1005). This necessitates further discussion on the provenance and origin of these raw materials.

No clear correlation between the amount of white infill and either the provenance of the sherd (e.g. Skorba or Santa Verna) or the type of decoration (e.g. chevron, line of C, net, cardial) was made. This conclusion is based on observations such as, for example, G1002 (Skorba, chevron) having minimal infill, while G1021 (also Skorba, but with 'Line of C' decoration) having a substantial amount. Similarly, G1004 (Santa Verna, net decoration) had significant infill, while G1005 (Santa Verna, cardial decoration) had minimal infill. These contrasting examples within each category suggest the absence of a link between the white infills and these factors based on this sample set (five samples).

A possible reason for the variation in the quantity of white infill observed among the analysed sherds can be attributed to post-depositional processes affecting the preservation of the white-infilled decorations. It can be assumed that during their manufacture, all sherds included in this research project featured decorations which were entirely infilled with a white material. However, over time, various post-depositional processes, such as abrasion, chemical weathering and biological activity, among other factors, could have led to the removal of differing quantities of white infill from these sherds (Gilstrap *et al* 2021: 6). These post-depositional processes can act selectively, affecting sherds in different ways based on factors such as their burial environment, exposure to environmental agents and inherent material properties (Gilstrap *et al* 2021: 6).

5.3 Provenance of the Raw Materials used in the White Infills

This section will consider the various minerals identified in the analysed white-infilled decorations, describing potential sources for each mineral.

Calcite

Calcite, a rock-forming mineral, is the predominant mineral found in all of the analysed samples, is commonly encountered in geological formations such as limestone, chalk, marble and marl. It also occurs in carbonate neo-formations and is a constituent of the shells of terrestrial and aquatic molluscs (Opriş *et al* 2022: 10). Provenancing the exact origin of the calcite used for the white infills in this study poses significant challenges due to the absence of comprehensive reference data, including rare-earth element (REE) data, even more so, since it can be found in a vast number of sources. Previous research has shed light on the distribution of calcite in both marine and terrestrial areas within the Maltese Islands, providing valuable insights into potential sources. The primary source of calcite-bearing minerals available to past and current societies are the Maltese limestone themselves, as limestone is predominantly composed of calcite (Bianco 1993, Bianco 2017: 4, Šiler *et al* 2018: 1). Given the abundance of limestone resources in the Maltese islands and the prevalent use of a white calcite-based infill for decorating pottery during the Għar Dalam phase, it is reasonable to conclude that Maltese limestone would be a primary candidate material potentially utilised in the production of these infills. However, an interesting question arises regarding the specific type of Maltese limestone employed for such purposes, as three distinct types including, Lower Coralline Limestone, Globigerina Limestone and Upper Coralline Limestone, can be identified as potential sources for the white infills. These three layers are themselves composed of different members (ERA n.d.), which could display a variation of mineral composition. The presence of *Globigerina sp.* in the white infill of G1030 suggests a potential association with Globigerina Limestone, as this limestone primarily consists of the tests of Globigerina foraminifera. In spite of this, one cannot easily disprove the potential of other Maltese sedimentary rock types as candidate materials, since planktonic foraminifera (*Globigerina*) have also been documented within these formations (Said 2011). Furthermore, it is possible that the foraminifera noted in the white infill infiltrated during the sourcing of clay or materials used for the white infills. Indeed, planktonic foraminifera have been found in local archaeological pottery and were very likely part of the raw material itself (Anastasi *et al* 2021). Moreover, considering that Globigerina Limestone covers approximately 70% of the superficial area of the Maltese Islands (ERA n.d., Bianco 2017: 3), and is the softest among the limestone varieties found on the islands (Said 2011: 14), it is reasonable to suggest its use for producing white infills. This characteristic of being relatively soft would make it easier to crush into a paste, facilitating its application in pottery decoration. Furthermore, certain members within the Globigerina

Limestone layer, such as the *Soll* type are softer compared to others like the *Franka* type (local terms) (Diana et al 2014: 251), potentially making these softer deposits better candidates for white infills if potters prioritised the ease of stone workability.

Another primary candidate material for the presence of calcite in the white infills is calcite crystal. This material is considered because it was noted in the present research project that calcite was added as temper (based on Santacreu (2014: 38)) for the clay used in the manufacture of G1030 (**Figure 57**), potentially suggesting its use in the white infills as well. This material has been documented in, for example, Maltese cemented fault rock resulting from diagenetic processes (Michie 2015). Furthermore, calcite veins within limestone formations are documented in the Maltese islands, exemplified by a megalith potentially placed deliberately at the entrance of the southern structure of Mnajdra (Wagner 2007: 75).

Calcite mineral may have also originated from marine-related sources. Turi *et al* (1990) conducted a study analysing the mineralogy of sediments from carbonate beaches across Malta (9) and Gozo (4) using XRD. Their research findings indicated that the majority of beach sediments are primarily composed of carbonates, with more than 90% originating from organic sources. Calcite was identified as the predominant mineral component, with a considerable portion stemming from the erosion of Tertiary formations. Additionally, they (Turi *et al* 1990: 373-374) linked the presence of high-Mg calcite in these sediments to recent porcelaneous foraminifera tests, red algae and echinoid remains and they noted that the $MgCO_3$ content in the high-Mg calcite was similar to that observed in recent carbonate sediments from the Mediterranean. The utilisation of primarily beach sediments in white infills appears improbable due to their natural coloration, which tends towards yellowish rather than white unless supplemented with additional whitening agents. Despite this, beach sediments as potential sources of calcite in the white infills were included in this section for the sake of comprehensive analysis, especially since the white infill of G1030 exhibited both white and yellow colourations under optical microscopy and also because aragonite, a mineral generally connected to beach sources was detected in one white infill (G1002). See section on aragonite below. Lastly, experimental archaeology could shed light on whether the mentioned calcite sources (e.g. different limestone types, calcite crystal sources or marine sediments) were feasible materials for creating the white infills observed in the pottery samples. By replicating the potential techniques and processes used, experimental archaeology can provide insights into the practical aspects of calcite processing and application in the white infills.

Quartz

Quartz, a hard and crystalline mineral composed of silica (SiO_2), ranks as the second most abundant mineral in Earth's continental crust, following feldspar (Anderson and Anderson 2010: 187). In mineralogical studies of Maltese limestone (Bianco 1993, Bianco 2017), the presence of calcite and quartz were consistently detected in all analysed samples (Murray 1890), suggesting a consistent association whenever calcite was present. Moreover, Quartz is a frequently found component of sedimentary rocks (Götze *et al* 2021: 639). Therefore, its presence in all of the white infills does not offer any indicative clues regarding the provenance of the materials utilised in their creation. A potential specific source for the presence of quartz in the white-infilled decorations, within the context of this study, may be chert, given that they it is primarily composed of quartz (Cilia 2010: 145). It is possible that inadvertent inclusion of chert fragments could have infiltrated the decorations, thereby leading to the detection of quartz in the white infills. Alternatively, there could have been a deliberate inclusion of crushed chert in the materials. However, this study did not uncover any evidence supporting the inclusion of chert in the white infill mixtures. Therefore, it is more likely that the quartz content resulted from its inclusion within other materials (such as limestone or clay sediments) used for the creation of the pastes.

Aragonite

Aragonite has traditionally been recognised as a marine carbonate (Bathurst 1968, Friedman and O'Neil 1977) and studies conducted in the Maltese islands have identified its presence in Maltese beaches. Turi *et al* (1990: 373) had detected minor quantities of aragonite in their analysis of the carbonate sediments from sandy beaches in Malta and Gozo, attributing its presence primarily to recent mollusc shells. Considering this, shells could have also been crushed and used to create the white infill. Aragonite sand seems to be more prevalent along the coasts characterised by Globigerina Limestone compared to high-Mg calcite sand, with the exception of areas where steep slopes of the Blue Clay Formation directly overlay these coasts in Northern Gozo and certain parts of Western Malta (Gatt 2007: 125). In these specific coastal regions, the occurrence of aragonite sand on beaches is diminished. Considering the information presented here, it is possible that materials (i.e. shells, sand and limestone) sourced from Maltese beaches were integrated into the production of the white infill observed in G1002. Potentially, it could be the case that during the procurement of clay used to create this sherd, the potters had also gathered the materials for the white infill. This hypothesis arises from the

fact that aragonite, a marine-related mineral, was found in the white infill and Skorba, where the sherd was found, lies approximately 4 km away from the nearest bays: Ġnejna Bay and Għajn Tuffieħa (**Figure 65**), both known for their clay deposits.
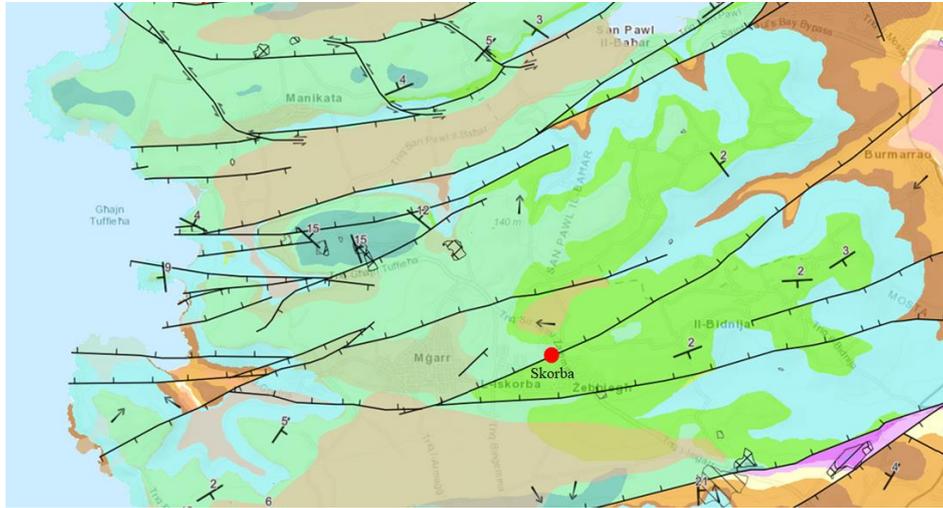
Figure 65: Figure illustrating clay deposits (light blue) in Ġnejna Bay and Għajn Tuffieħa, both of which are in the vicinity of Skorba (red dot). Image adapted from the Geological Map of the Maltese Islands (Continental Shelf n.d.).

Due to this the following questions have been raised:

1) Was the material, specifically the clay or the white infill components (or potentially both), sourced from a nearby bay like Ġnejna Bay or Għajn Tuffieħa, and then manufactured in the vicinity of Skorba?

2) Was the material sourced from a distant coastal area and transported to the vicinity of Skorba for manufacturing?

3) Alternatively, was the sherd manufactured elsewhere and subsequently deposited at the site of Skorba?

The questions posed above are challenging to address within the scope of the present study, especially due to the absence of comprehensive published reference data.

While aragonite is primarily associated with marine environments, caves serve as an alternative source for its presence. Aragonite formation is widespread in limestone caves globally and is commonly found in cave formations such as stalactites and stalagmites. Speleothems can be composed of aragonite, but aragonitic speleothems are primarily found in caves formed in dolostone or dolomitic limestone (Rossi and Lozano 2016: 71). Although dolomitic limestone is not readily found on the Maltese islands, investigating the potential

presence of aragonite in Maltese speleothems, such as those in Għar Dalam Cave, is a possibility. Mineralogical analysis would be necessary to confirm whether aragonite is indeed present in Maltese speleothems and could have been used as materials for the white-infilled decorations. However, this hypothesis appears less probable, particularly due to the relative rarity of speleothems, in contrast to the prevalent practice of embellishing pottery decorations with a white material during the Għar Dalam phase. This would only be more likely if there was a specific cultural significance associated with using speleothems for creating white infills. Nonetheless, this hypothesis cannot be easily dismissed, as Għar Dalam phase pottery has been discovered in cave contexts (e.g., Għar Abdul, Għar Dalam, Għar ta' Għeju) (**Figure 3**). Based on the evidence presented, it is more probable that the materials used for the creation of G1002 were acquired from coastal regions, where shells and other marine materials containing aragonite are present.

Fluorapatite

Fluorapatite, a phosphate mineral known for its hardness and crystalline structure, was identified within one of the white infills (G1005). An intriguing source for the presence of fluorapatite in this context could be the utilisation of shark and other fish species' teeth. Unlike mammals, which primarily utilise hydroxyapatite, fish incorporate fluorapatite into their tooth biomineral (Enax *et al* 2012: 299). The use of shark teeth for incising clay in Maltese Neolithic pottery production has been documented (Evans 1971: 153, Sagona 2015: 144, Malone *et al* 2020a: 349). It is conceivable that during the creation of the pottery decorations, shark or fish teeth fragments could have inadvertently infiltrated the white infill mixture. This would suggest that materials close to beach sediments were utilised. If the argument for the proximity of an archaeological site to the nearest bays with clay is applied here as was done for Skorba, the closest marine sources to Santa Verna would be the coastline from Ramla Bay to Marsalforn Bay, ranging from approximately 2.5 to 2.8 km away. Alternatively, ancient potters may have intentionally integrated shark or fish teeth into the white infill mixture, but if this was the case, it would raise inquiries on the reasons for doing this.

The detection of fluorapatite might also be linked to the presence of francolite in the white infill, which is a type of carbonate fluorapatite formed through microbial precipitation and characterised by its low crystalline structure (Soldati *et al* 2010: 10). Francolite serves as a significant component of phosphorites, sedimentary rocks rich in phosphate content, typically ranging from 18-40 wt% P_2O_5. Within the francolite structure, the CO_3^- group is believed to

substitute for PO$_4^-$ groups, forming an integral part of the fluorapatite composition. In Malta, francolite is mainly found within the phosphorite beds found across the Maltese Islands (Pedley and Bennett 1985: 18), with the most pronounced occurrences in the western and northern regions (Carbone *et al* 1987: 35). These beds span from the Aquitanian (*c*. 23-20 mya) to Late Burdigalian (*c*. 20-15 mya) epochs. Such phosphate conglomerate beds serve as distinct stratigraphic markers within the Globigerina Limestone sequence (Cilia 2010: 124). Designated as C1 and C2 in Cilia's work, respectively, he mentions that they demarcate the boundaries between the Lower and Middle layers, and the Middle and Upper layers. Furthermore, such beds, comprising phosphatised pebbles and botryoidal nodules, often house an abundance of phosphatised fossils and exhibit distinct characteristics. Interestingly, layers of brown phosphatic conglomerates are found at the base of the Upper Globigerina Limestone layers along the coastline from Ramla Bay to Marsalforn Bay (marked with dark brown in **Figure 66**) (Continental Shelf n.d.). This observation suggests that the fluorapatite detected in G1030 may have originated from these beds, acquired either intentionally or inadvertently during the collection of white infill material (e.g. Globigerina Limestone) or clay (marked as light blue in **Figure 66**). Determining whether the source of the fluorapatite within the white infill was from these phosphate conglomerate beds or shark teeth poses a significant challenge due to the lack of reference data for comparison. Moreover, for such a study to yield meaningful results, it is imperative that additional white-infilled decorations are found to contain a presence of fluorapatite. This requirement arises because, in the present study, only one decoration has been shown to contain a significant amount of phosphorus.
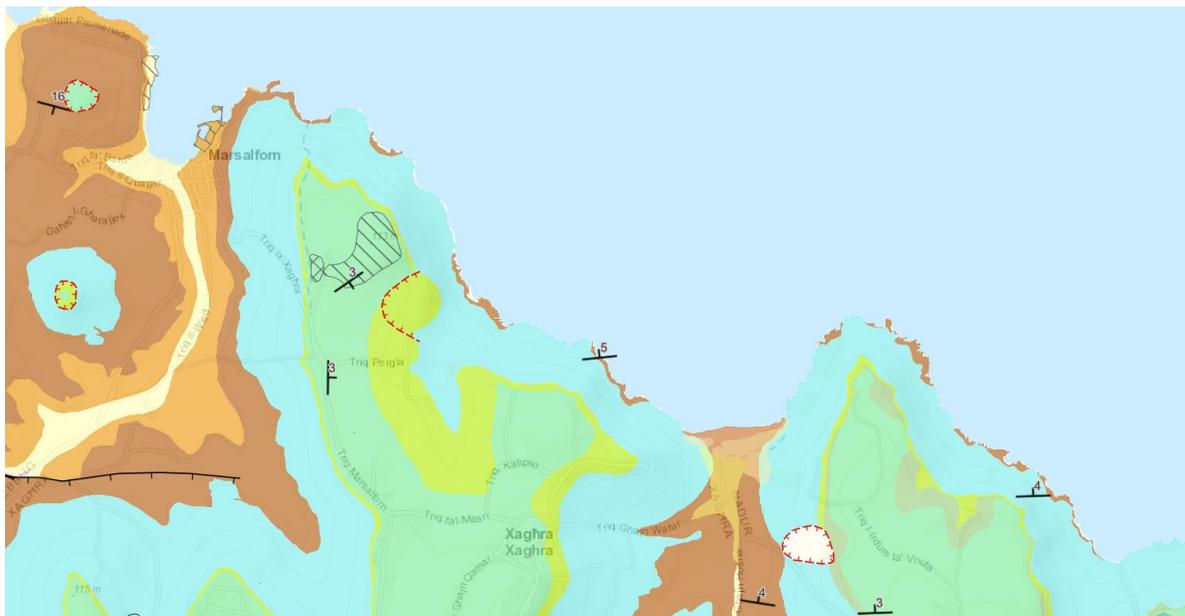
Figure 66: Geological map focusing on the coastline from Ramla Bay to Marsalforn Bay (Continental Shelf n.d.). Light blue marks the clay sources and the dark brown mark the Upper Globigerina Limestone sources, with brown phosphatic conglomerate layers found underneath (Continental Shelf n.d.)

Dolomite

Dolomite crystallises in the rhombohedral system and is distinguished from calcite by the inclusion of magnesium ions, altering its chemical formula to $CaMg(CO_3)_2$. While dolomite is a prevalent sedimentary rock-forming mineral, often found in massive beds spanning several hundred feet (Soldati *et al* 2010: 21), it is not as abundant in the Maltese islands. In fact, large dolomite outcrops are absent in the Maltese islands, yet its presence in the white infills can be attributed to various potential sources. As previously mentioned in **Section 2.1.4**, dolomite in Malta is situated beneath observable rock layers in the Maltese Islands, which comprise multiple strata of sedimentary rocks dating back to the late Triassic period (Cilia 2010: 116). Obtaining dolomite from this source appears unlikely due to the impossibility of sourcing materials from such depths, especially considering the discovery of dolomite in three separate white infills. The Blue Clay stratum also serves as one potential source of dolomite in the Maltese islands, as it occasionally contains this non-clay mineral (Soldati *et al* 2010: 20). Furthermore, dolomite has been observed within the Globigerina limestone layer, specifically in the "terminal Middle Globigerina Limestone Omissionground" (TMGLOg) and "Fomm-ir-Rih Local Hardground" (FiRLHg) surfaces (Gruszczyński *et al* 2008: 239). Lithification occurred in two phases, with dolomite believed to have formed below the seafloor. Subsequent

exhumation led to partial corrosion, coinciding with the precipitation of calcitic and phosphatic cements in burrows exposed to seawater circulation.

The presence of dolomite may also be attributed to the existence of high-Mg calcite. It is documented that the precipitation of high-Mg calcite could serve as one of the precursors to dolomite formation (Gregg *et al* 2015, Rodriguez-Blanco *et al* 2015, Hobbs and Xu 2020). Furthermore, distinguishing between high-Mg calcite and dolomite XRD can be challenging. High-Mg calcite can exhibit a 104 peak at the position typically associated with dolomite, leading to potential misidentification (Kaczmarek *et al* 2017: 1-3). True dolomite is characterised by additional ordering reflections which are absent in high-Mg calcite (**Figure 67**) (Kaczmarek *et al* 2017: 3). Thus, the presence of dolomite-specific ordering reflections is essential for accurate identification, ensuring that the observed peaks are not solely due to high-Mg calcite. Dolomite was confirmed in this research project by comparing the XRPD patterns from Kaczmarek *et al* (2017) (**Figure 67**) with the XRD data generated in this study.
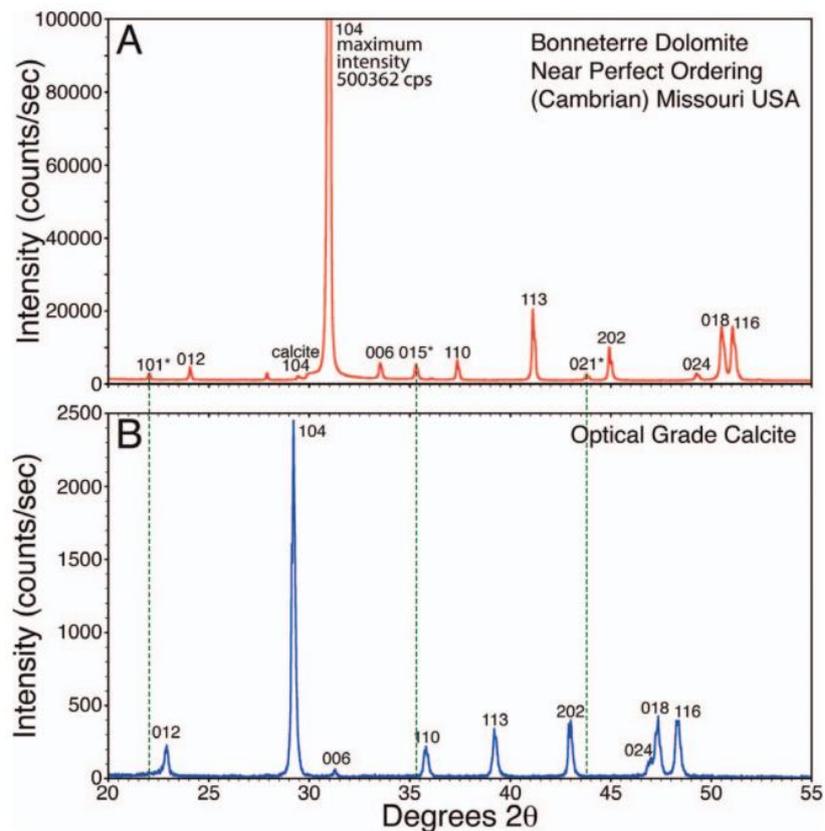
Figure 67: Comparison of XRPD patterns of dolomite (A) and calcite (B) from 20 to 55° 2θ (Kaczmarek *et al* 2017: 3)

Additionally, certain hypotheses suggest that foraminifera could contribute to the detection of dolomite. Zhao *et al* (2024) observed dolomite crystals within planktonic foraminifera (i.e. *Globigerinoides sp.*) through SEM, XRD and stable isotope analysis. They (Zhao *et al* 2024: 1) hypothesised that high-Mg calcite may serve as the precursor for foraminiferal dolomite, with dissolution-precipitation processes potentially playing a role in the ageing process of dolomite formation. Planktonic foraminifera, particularly those belonging to the *Globigerinidae* family, are prominently observed on the Maltese islands, as highlighted in **Section 2.1.4**, so much so that a specimen from the *Globigerinidae* family was discovered in one of the white infills (**Figure 51**), despite the relatively low number (5) of samples analysed. Given these observations, it is plausible that foraminifera tests served as the source for the detection of dolomite in the white infills. Additionally, other organisms such as calcareous red algae and echinoids, which contain high-Mg calcite and are present in Maltese geology as highlighted in **Section 2.1.4**, could also have contributed to the detection of dolomite in the white infills. As demonstrated in this section, high-Mg calcite has the potential to lead to the detection of dolomite in micro-XRD analysis.

Considering the relationship between the Għar Dalam phase people and Sicily and Italy, it is worth noting the abundant presence of dolomite in the Italian region. For example, the Dolomite Mountains, situated in northeastern Italy, derive their name from their primary composition of dolomite (McKenzie and Vasconcelos, 2009). Closer to the Maltese islands, large dolomite outcrops have also been identified in Sicily (Di Bella *et al* 2020). In spite of the fact that, dolomite is more readily available in Italy and Sicily due to the presence of extensive outcrops and its occurrence in Malta is limited, the present study refrains from suggesting that the dolomite found in the white infill originated from foreign sources, as there is insufficient evidence to support such a claim. However, this hypothesis remains a possibility that requires further scientific evidence to substantiate.

The following table (**Table 17**) outlines the minerals identified in the white infills with their potential sources displayed in brackets.

Mineral	Potential Sources
Calcite	Maltese Limestone (Lower Coralline, Globigerina, Upper Coralline), calcite crystal, marine sources (foraminifera tests, beach sediments)
Quartz	Common mineral in all of the Maltese sedimentary layers, Chert
Aragonite	Marine sources (shells, beach sediments), Caves (speleothems)
Fluorapatite	Marine sources (shark and fish teeth), Phosphorite beds
Dolomite	Limestone layers, Blue Clay, foraminifera (*Globigerinoides* sp.), high-Mg calcite, foreign material (?)

Table 17: Potential sources for the minerals found in the white infills

Phosphate Mineral Fragment

This section discusses the fragment (**Figure 58**) identified within the white infill of G1030. SEM imaging of this fragment did not reveal distinctive characteristics comparable to known mineral SEM images at the analysed scale, precluding direct comparison for identification. However, EDS analysis utilising the point function indicated a predominant presence of a mineral rich in calcium and phosphorus, suggesting classification as a phosphate mineral. This classification encompasses hydroxyapatite ($Ca_5(PO_4)_3OH$), fluorapatite ($Ca_5(PO_4)_3F$) or chlorapatite ($Ca_5(PO_4)_3Cl$). Chlorapatite was excluded from consideration due to the absence of detected chlorine. Considering the mineral composition, the fragment is likely composed of either hydroxyapatite or fluorapatite. Despite the relatively low fluorine content in the fragment, fluorapatite remains a possibility. This is because EDS analysis (**Figure 68**) demonstrates that fluorapatite typically exhibits lower fluorine content in comparison to calcium and phosphorus.
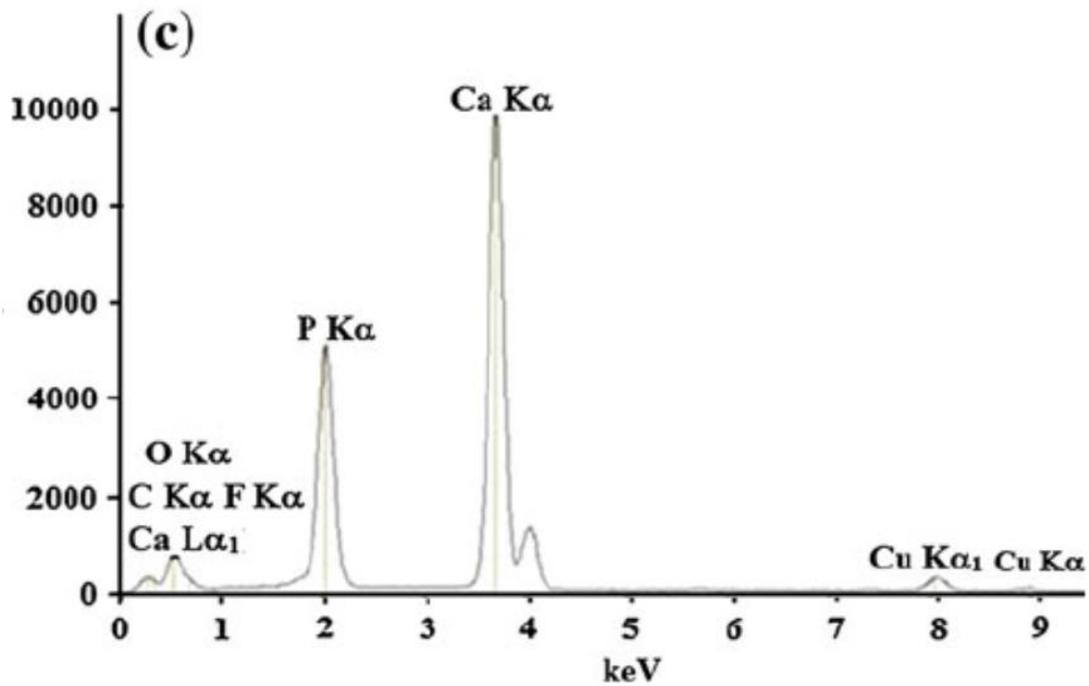
Figure 68: EDS analysis of fluorapatite (Iqbal *et al* 2013: 77)

Furthermore, the presence of fluorapatite would not be surprising, as it was previously detected through micro-XRD in another white infill (G1005). While the possibility of the fragment being hydroxyapatite cannot be disregarded, since bone seems to have been a common mineral used in the production of prehistoric white infills (Odriozola *et al* 2012: 149, Giustetto *et al* 2013: 4254, Opriş *et al* 2022: 8), the absence of detected hydroxyapatite or fluorapatite in the micro-XRD analysis of G1030 precludes definitive conclusion regarding its mineral composition.

As a summary, the following table (**Table 18**) presents the minerals found in the white infills analysed in the present study. In the next section, the mineralogical data will be compared with other scientifically analysed white infills from the Central Mediterranean to determine similarities and/or differences.

Sample	Decoration	Minerals Identified	Archaeological Site
G1002	Chevrons	Calcite, Quartz, Aragonite	Skorba
G1004	Net	Calcite, Quartz, Dolomite	Santa Verna
G1005	Cardial	Calcite, Quartz, Dolomite, Fluorapatite	Santa Verna
G1021	Line of 'C' Pattern	Calcite, Quartz, Dolomite	Skorba
G1030	Band of Diagonal Lines	Calcite, Quartz	Santa Verna

Table 18: Minerals identified in the white infills

5.4 Comparison with Central Mediterranean White-infilled Decorations

This section will compare previous data on Central Mediterranean white-infilled decorations with the results of the present study to elucidate implications from the observed similarities and differences. Scarcella (2011) reports that the analysed Stentinello white-infilled decorations from Capo Alfiere and Perriere Sottano, as well as an Għar Dalam phase sherd from Skorba, exhibited a calcium-based composition. This finding is consistent with the results of the present study, which also identified a calcium-based composition in the Għar Dalam phase white-infilled decorations. The sole XRD analysis conducted by Scarcella (2011) on a white-infilled impressed line decoration (PER7) revealed the presence of calcite and quartz. Similarly, the present study found these minerals in all the analysed samples. Unfortunately, the white infill in the Skorba sherd was not subjected to mineralogical analysis, preventing a direct comparison with the current study's findings. Additionally, Scarcella (2011) notes variations in the application of coloured infills. However, macroscopic and microscopic examination of the white infills in the current study did not observe such variations. Instead, the white infills were consistently applied within the contours of the impressions or incisions on the sherds.

The study conducted by Giustetto *et al* (2013) yielded results that diverged from those of the present study. The sherds analysed by Giustetto *et al* were from Piedmont, northwestern Italy, distinguishing them from the sherds analysed by Scarcella (southern Italy and Sicily) and

the present study (Malta and Gozo). Giustetto *et al* determined that the main minerals used for the white infills included talc and bone, which were the most common, and sporadic kaolinite. In contrast, the present study did not find conclusive evidence for any of the minerals identified in the study by Giustetto *et al*, indicating variations in materials used and in the *Chaîne Opératoire* from white infills studied by Scarcella (2011) and the ones analysed in the present study. Reasons for this variation could be geographical. Talc is abundant in large outcrops in Piedmont, as noted by Giustetto *et al* (2013: 4257), making it an accessible material for the white infills, while the use of bone may could potentially be attributed to the region's mountainous geography, which likely supported a larger population of large mammals, thereby providing more bones for white infill creation.

5.5 Manufacturing Techniques

This section will discuss the insights into manufacturing techniques that can be derived from the elements and minerals present in the white infills. The white infills observed in the samples were predominantly composed of calcite, serving as the primary whitening minerals in the white infill mixtures, with low amounts of quartz. These minerals were likely obtained through the crushing and grinding of local carbonate sources, such as Maltese limestone and calcite crystals. Other minerals resulted either through their conscious inclusion or through infiltration during the procurement of raw materials or the pottery production process.

Firing

Insights into the firing techniques employed by ancient potters can also be obtained from the examination of the data gathered for this research project. Research on the effects of firing on calcitic phases shows that calcite starts to decompose at 700°C and largely disappears by 800°C (Cultrone *et al* 2001: 624). Nonetheless, traces of calcite can still be found at 800°C (Cultrone *et al* 2001: 633). The presence of calcium-based inclusions in the ceramic matrices (observable in the SEM images) and the unaltered state of the white infills suggest that the firing temperatures used in producing the sherds examined in this research project likely did not exceed 800°C. This indicates a relatively low firing temperature, consistent with the conclusions drawn by Molitor (1988: 204), who suggested temperatures below 980°C. Based on the evidence, it is plausible that the firing temperature was below 800°C. Due to the low firing temperatures used and the methods employed in this study, it is not possible to determine whether the white paste was applied before or after firing, as the minerals were not

thermodynamically affected, which prevents distinguishing between pre- and post-firing application.

Binders

The examination of the white-infilled decorations did not detect the presence of any binders to secure the white paste to the decorations. This absence may be attributed to the lack of specific analytical techniques utilised for the detection of binders, particularly those potentially of inorganic as noted by Scarcella (2011: 208-209) or organic origin as suggested by Opriș *et al* (2022), or because the binder had decomposed due to firing. However, it is conceivable and probable that binders, such as clay, water, egg, milk or plant-based residues which were available to the people of the time, were utilised in the creation of Għar Dalam phase pottery white-infilled decorations.

Possible Chaîne Opératoire

This section presents a possible *chaîne opératoire*, which appears to be the most probable method for creating the white-infilled decorations in Għar Dalam phase pottery. Firstly, clay would likely be sourced from local deposits, potentially from nearby clay sources like Ġnejna Bay for pottery sherds found at Skorba, or nearby clay sources for the sherds found at Santa Verna. The clay would then undergo typical preparation techniques possibly including the following steps: sorting or levigation, temper inclusion, wedging and kneading (Borg 2005). Then, the forming of the pot could include pinching and drawing, slab building, coiling and moulding (Borg 2005). Surface treatment would follow and this could include the following: beating, scraping, smoothing, burnishing, polishing, texturising and decorating (Borg 2005). In the case of Għar Dalam phase pottery, burnishing and incision and impression tools like sticks, bones, shells or fingernails would be possible. Afterwards, the pot would be dried and then fired, transforming the malleable clay into pottery (Borg 2005).

If the white paste was applied before firing, the materials used for its creation, likely Maltese limestone or calcite crystal, possibly containing minerals such as aragonite, fluorapatite, quartz and/or dolomite, would have been gathered beforehand. These materials would then be finely crushed into a paste. Afterwards, a binder such as clay, water, egg, milk or plant-residue would be applied either to the impressed decorations, the crushed materials or both. This would allow the fine paste to adhere to the impressions. The vessel with the white-infilled decorations would then be fired at a relatively low temperature (<800°C). If the white paste was applied after firing, the manufacturing process, application techniques and firing

temperature would remain the same. The distinction lies in the timing of application, which occurs post-firing rather than before. While this suggested *chaîne opératoire* does not aim for complete accuracy and variations were likely present during the Għar Dalam phase, summarising a possible *chaîne opératoire* in this section provides the benefit of visualising how the creation of white-infilled decorations may have been carried out.

Implications on Technological Knowledge

The present study offers insights into specific implications of the technological knowledge possessed by ancient potters responsible for creating the earliest white-infilled decorations in Malta. While previous research has addressed aspects such as pottery production techniques and the creation of impressed decorations in the Għar Dalam phase (Evans 1954, Trump 1966, Molitor 1988, Pirone 2017, Vella Gregory 2021, Richard-Trémeau *et al* 2023a), this section will focus primarily on the technological knowledge associated with the creation and application of the white infill.

Firstly, potters who crafted Għar Dalam phase white-infilled decorations possessed an understanding of the local materials required for their production. While white-infilled decorations in Europe appear to have spread from East to West, coinciding with the dissemination of the Impressed Ware culture, the primary materials utilised for creating the white pigments varied significantly due to regional resource availability. For instance, while the Vinča culture in the Balkans favoured the use of bones for white-infilled decorations, the Għar Dalam people seem to have predominantly relied on locally sourced limestones or calcite crystal, although there is potential evidence of bone utilisation in one sample (G1030). One possible explanation for limestone use could be that the potters were aware of the accessibility and workability of Maltese limestones, particularly the Globigerina type. This would have facilitated the procurement and crushing process. An explanation for the use of calcite crystals could be inferred from the fact that potters utilised this material as temper in sherd (G1030); hence, it is plausible they also used it for the white infills. In addition, the use of limestone or calcite crystals may have been influenced by specific cultural or practical considerations among the potters. However, this detail will only be mentioned in the present study without further exploration, as it is beyond the scope of this research project. Lastly, while conclusive evidence supporting the use of bones in the white infills has not been found, it cannot be definitively concluded that bones were not used for several reasons: Firstly, a fragment potentially being bone or fluorapatite was observed in one white infill, although its identification remains

inconclusive. Secondly, the sample size (5) is too small to assert that bones were never used. Thirdly, bone use was prevalent in other impressed white infills elsewhere, suggesting it could have been utilised in the Għar Dalam phase as well. Finally, it is plausible to suggest that bones could have been used in the white infills, as they could have been readily sourced from meals.

Another implication pertains to the Neolithic peoples' understanding of adhesives and binders in pottery decoration. This research project posits that a binder was likely employed to adhere the white paste to the impressed decorations, despite the lack of scientific evidence for its presence. This hypothesis arises from the belief that a binder would have been necessary to adhere the crushed materials to the impressions. Similarly, Opriș *et al* (2022: 12) also found no evidence for the use of binders but inferred their existence. It is evident that the artisans, either through trial and error or intergenerational transmission, recognised the necessity of applying a material, to the crushed white paste itself, the impressed decorations or both to affix it securely. Such materials may have included binders such as (water, egg, milk or plant-based residues), reflecting the technological sophistication and practical ingenuity of the Għar Dalam phase culture.

5.6 Function

While it is generally accepted that the primary purpose of Għar Dalam phase white-infilled decorations was to enhance the impressed decorations (Sagona 2015: 30), one may ponder why this was the case, considering the additional energy required for their manufacture. Could it have been solely for this reason, or might there have been other factors at play, such as artistic, social or religious motivations, prompting the use of white infills to adorn their pottery? The current study refrains from proposing a hypothesis on this matter as it is beyond the scope of this research project. However, this discussion is included to illuminate the reader, sparking curiosity about the possible multifaceted motivations behind the ancient white-infilled decorations. This inquiry gains particular significance when considering the local context, since the subsequent Grey Skorba phase exhibits "a complete absence of decoration" (Trump 1961: 301), while the Red Skorba phase features red slip as its primary decorative element, contrasting with Għar Dalam phase pottery decorations.

Chapter 6: Conclusion

6.1 Summary

Multi-analytical archaeometric analysis was conducted on five white infills inlaid on Early Neolithic impressed pottery sherds from the Għar Dalam phase, originating from the sites of Skorba and Santa Verna. Calcite emerged as the predominant decorating material, with additional minerals such as quartz, aragonite, dolomite and fluorapatite also present. Calcite and quartz were identified in all samples, forming the exclusive minerals comprising the white infill of sample G1030. In contrast, two samples, one from Skorba and the other from Santa Verna, exhibited mixtures of calcite, quartz and dolomite. Further variation was observed, with a white infill from Skorba composed of calcite, quartz and aragonite, while a sample from Santa Verna displayed a composition featuring calcite, quartz, fluorapatite and dolomite. The dominant presence of calcite (Maltese limestone or crystal calcite) as the primary whitening agent, with the likely infiltration of other minerals, suggests that a standardised mixture was employed in creating Għar Dalam phase white-infilled decorations. The use of local raw materials underscores the adaptability of the potters to their surroundings. However, deciphering the technology behind preparing and applying pottery pigments poses challenges due to uncertain material sources. Although no traces of binders were identified, further investigation using techniques such as, FTIR, Raman spectroscopy and chromatographic techniques, could potentially detect binders like water, egg, milk or plant residue on other pottery samples, thereby challenging this hypothesis. However, it remains uncertain whether such binders could be reliably distinguished from the bulk material. The findings made in this study refute the suggestion originally proposed by Trump (1966) that Għar Dalam phase white-infilled decorations were composed of gypsum.

6.2 Limitations of the Study

One limitation of the present study was the small number of white-infilled decorations (five) that were analysed. Incorporating a larger sample set could have revealed the presence of additional trace minerals, which could have potentially provided further insight into the provenance of the materials used or the manufacturing techniques employed. Despite the limited number of samples analysed, it appears that calcite was consistently used as the primary component in the white infills and further sample analysis would likely also show that this was the case. Another limitation of the current study was the restricted quantity of white infill

sampled. To mitigate this, we obtained an additional sample from G1004 for EDS analysis and another from G1004 for micro-XRD. The results indicated homogeneity in the white infill, however, this analysis was not conducted on multiple areas of all the samples, which would have been preferable.

6.3 Recommendations for further research

Finally, this research has yielded seven insights that warrant consideration for future studies. These insights are listed below:

i. Based on the findings of this research, additional archaeometric studies of other Għar Dalam phase pottery white-infilled decorations would likely also detect calcite as the main component, but additional scientific studies of the white infills could uncover the presence of distinct minerals in varying proportions or mixtures, which could provide further insights into the provenance of the materials used for the white infills and the manufacturing processes involved. Alternatively, analysis that can quantitatively determine the presence of trace elements would offer a more precise method for establishing the provenance of the materials used in the white infills. The results from such studies could also be compared with foreign analysed white-infilled decorations, namely Stentinello ware, which could yield new implications.

ii. Furthermore, conducting further archaeometric studies on white-infilled decorations from different phases in Maltese prehistory (i.e. Mġarr, Tarxien, Borġ in-Nadur; etc.) is crucial for enhancing our understanding of their composition. Given the limited scientific research on this subject, these studies could potentially reveal whether the similar recipe was consistently used for creating white-infilled decorations throughout Maltese history, suggesting a deliberate continuity in manufacturing practices. Alternatively, they may uncover variations in materials used, potentially offering insights into the provenance of these materials and the manufacturing techniques employed.

iii. A future study could involve conducting scientific analyses, such as LA-ICP-MS, to target and quantify trace elements in local carbonate materials from potential white infill sources, including limestone layers and calcite crystal sources on the Maltese islands. This reference data could be instrumental in determining the provenance of the calcite used in the white infills.

iv. Another avenue for research entails the scientific analysis of Maltese speleothems to ascertain their chemical and mineralogical composition, possibly identifying the presence of aragonite within them. Such analysis could shed light on whether these speleothems served as a potential source for the white infills in Għar Dalam phase pottery.

v. Further archaeometric analyses on Għar Dalam phase white-infilled decorations can help determine whether fluorapatite was consistently used in their creation. If fluorapatite is consistently found, further investigation could try to ascertain whether its source is local phosphate beds, such as along the coastline from Ramla Bay to Marsalforn Bay, shark/fish teeth or potentially some other source. Additionally, such a study could shed light on whether fluorapatite was consciously or inadvertently included in the white infills.

vi. The present study carried out chemical analysis on both the ceramic body and the white infill to clearly define the chemical variations between the two and to determine possible variations in the clay bodies (building upon previous studies including Evans 1954, Trump 1966, MaltaPot, etc.). In view of this, future research could concentrate on trying to identify the sources of clay utilised in the manufacture of Għar Dalam phase vessels. This is particularly interesting because the current study suggests a possible correlation, based mainly on the geographical proximity of potential candidate source materials, such as clay and white infill material, from near Ġnejna Bay and Għajn Tuffieħa Bay, which may have been used for producing the pottery found in Skorba. Similar correlations are noted along the coastline stretching from Ramla Bay to Marsalforn Bay, possibly pertaining to the Santa Verna pottery. The study would entail determining which local outcrops were utilised and investigating the potential use of foreign clays. Moreover, researchers could explore whether pottery production involved the use of single or multiple clay sources.

vii. Finally, experimental archaeology offers potential for understanding past technologies by attempting to recreate white-infilled decorations similar to those made by the Għar Dalam people. This methodology would entail replicating the entire pottery production process, commencing from sourcing clay for vessel construction to procuring materials for the white infills, with a priority for locally available resources. One aspect of this study would involve investigating the behavioural properties of the different types of Maltese limestones during crushing, using tools available to the people of the time. This investigation would aim to identify variations in methodologies and the resulting pastes, ultimately discerning which

closely resembles the white pastes used in Għar Dalam phase pottery. After the reconstructions are created, there is potential for conducting chemical and mineralogical analyses on these replicas which could reveal the presence of any unintended inclusion of additional minerals. The presence of unintended minerals in the reconstructions may imply that the low quantity minerals (i.e. quartz, dolomite, aragonite and fluorapatite) detected in the analysis of the white-infilled decorations in the present study were also unintentional and inadvertently gathered during the procurement of the calcite sources. Moreover, experimental archaeology provides an opportunity to investigate the need for binders in affixing the white paste to decorations. Reconstructions can attempt to create white-infilled decorations without utilising binders to assess the feasibility of this approach. Should this prove impractical, researchers could experiment with various binders accessible to ancient Għar Dalam phase inhabitants, such as clay, water, egg and milk, to try to determine the most effective options for this task. A potential limitation of conducting this experimental archaeology study in the Maltese Islands is the lack of dedicated facilities or spaces for experimental archaeology research. Such facilities would be instrumental for this project as it heavily relies on utilising local materials throughout the entire process to recreate the most accurate Għar Dalam phase pottery and white-infilled decorations.

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Appendix

A Electron Images
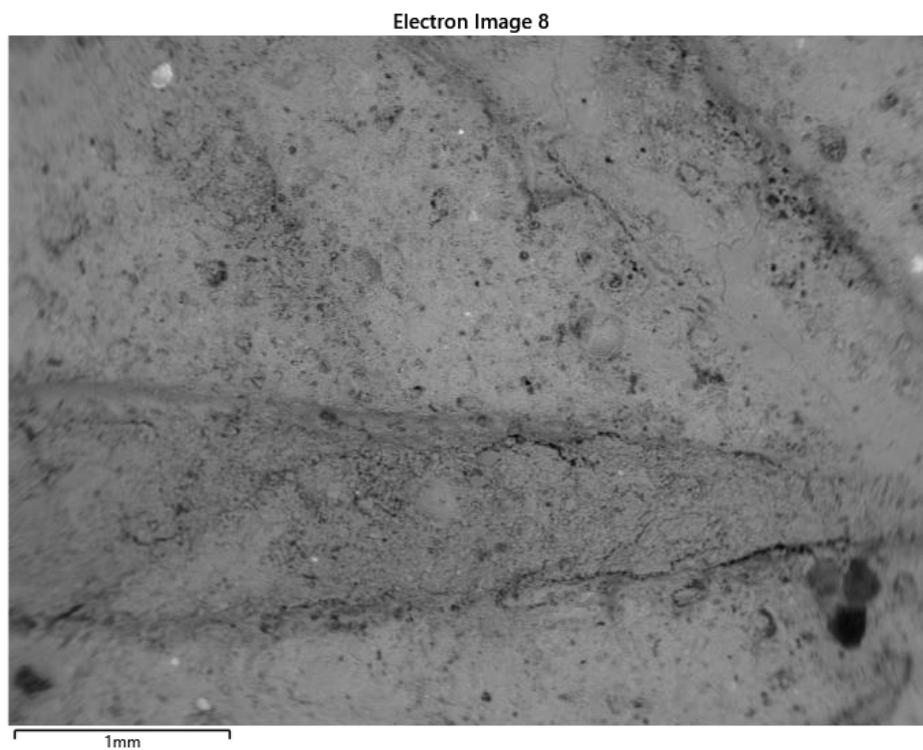
Figure 69: Electron image of the area analysed with EDS (G1002)
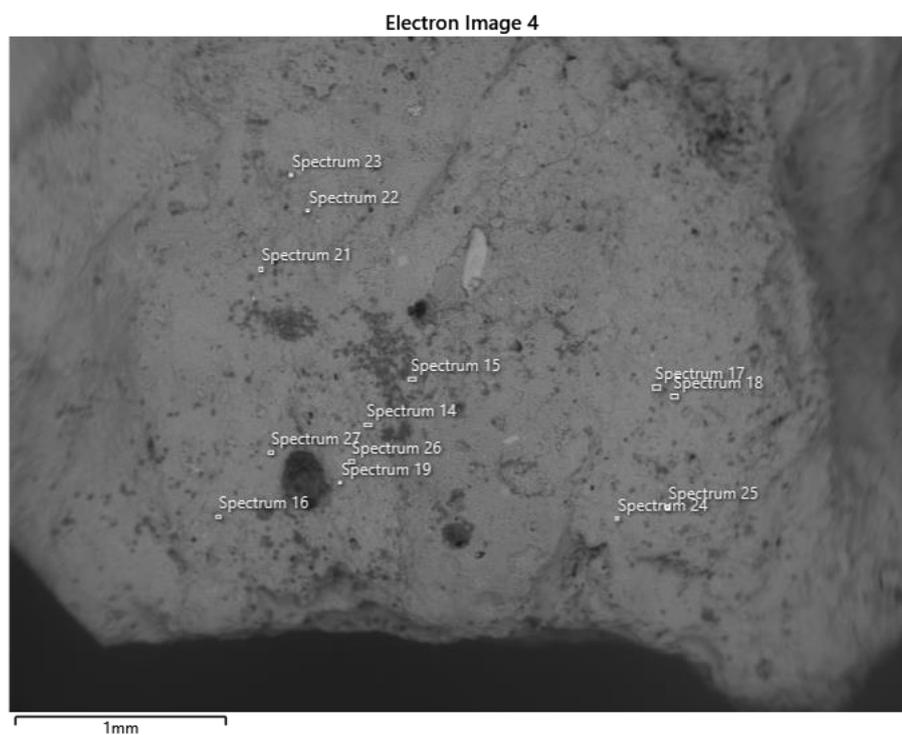
Figure 70: Electron image of the area analysed with EDS (G1004) (analysis no. 1)
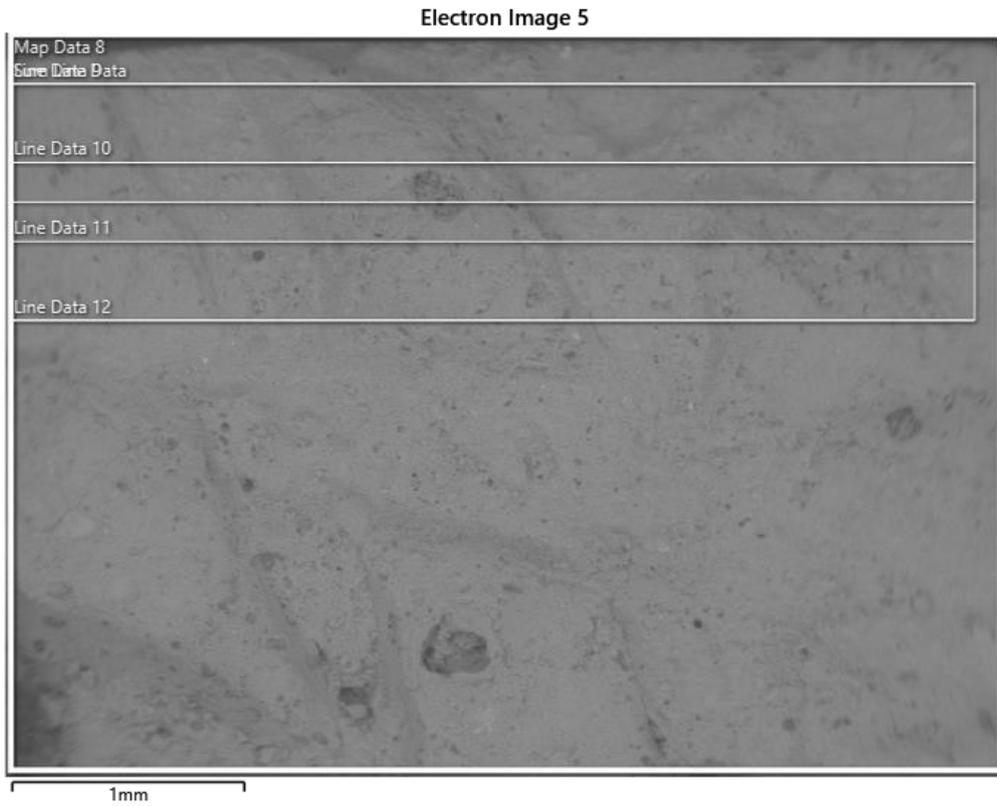
Figure 71: Electron image of the area analysed with EDS (G1004) (analysis no. 2)
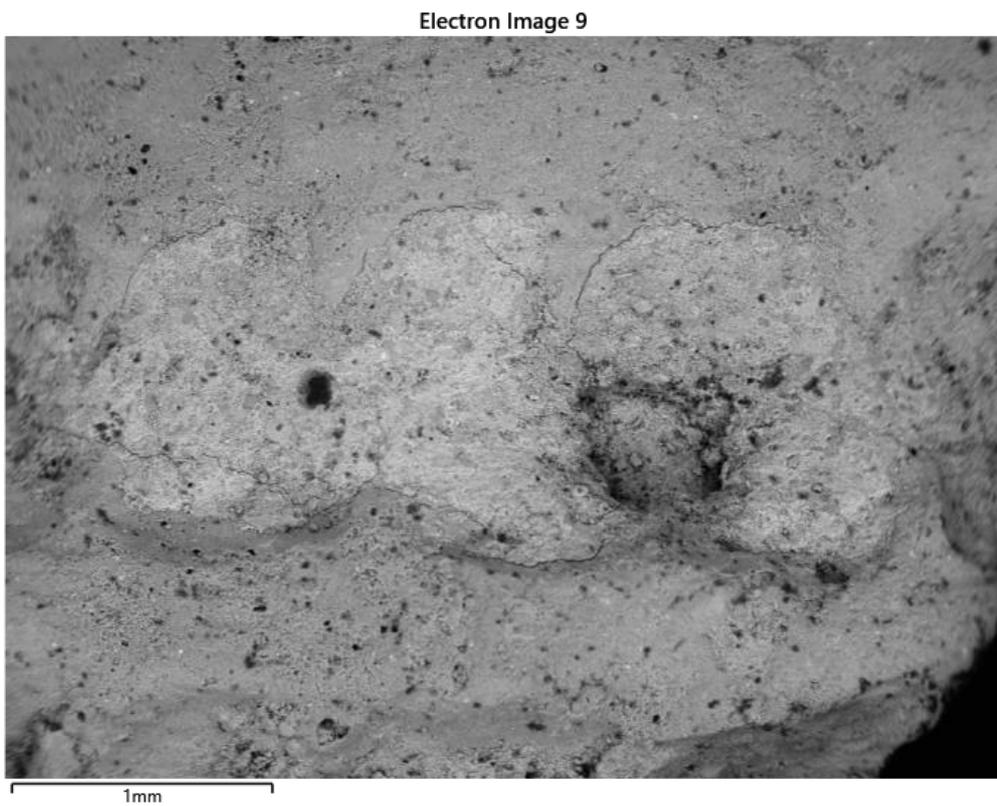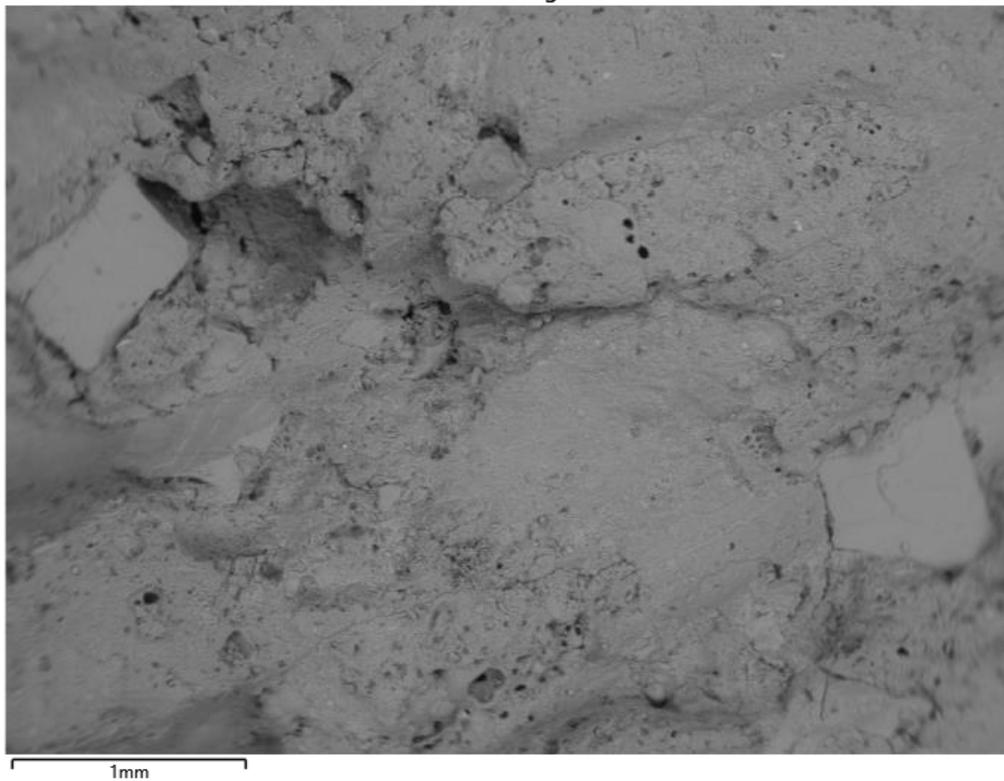

Figure 72: Electron image of the area analysed with EDS (G1005)

Electron Image 6
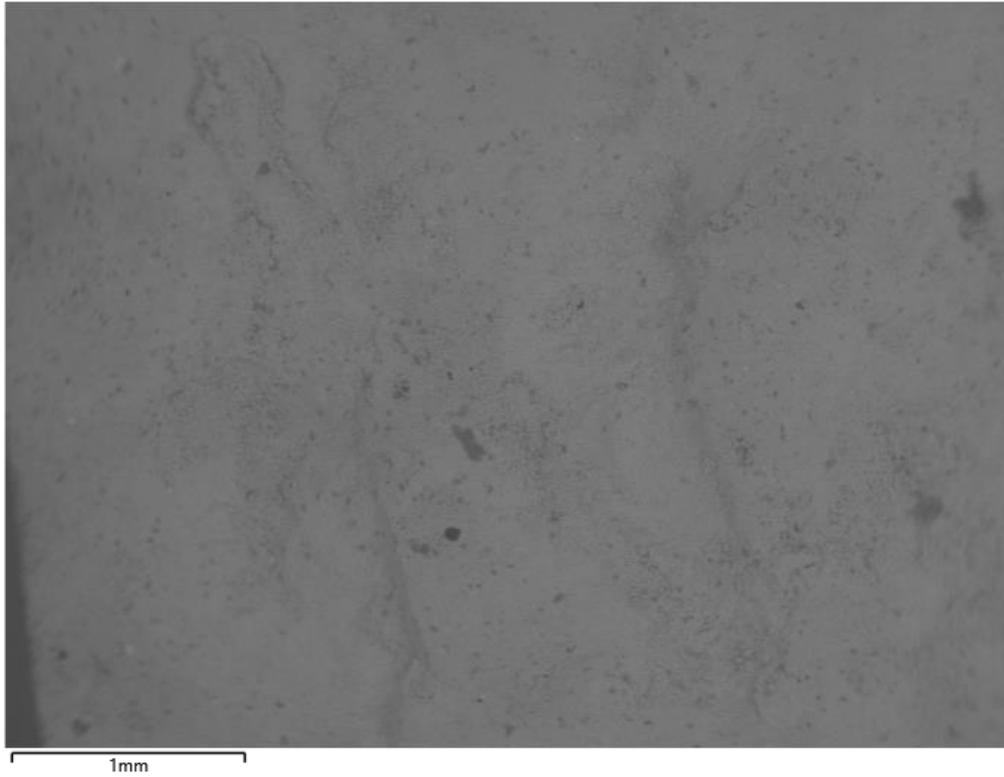
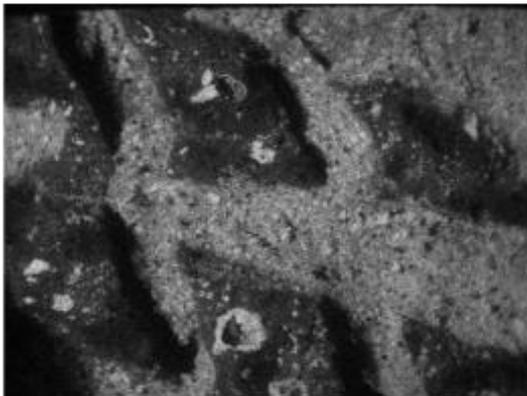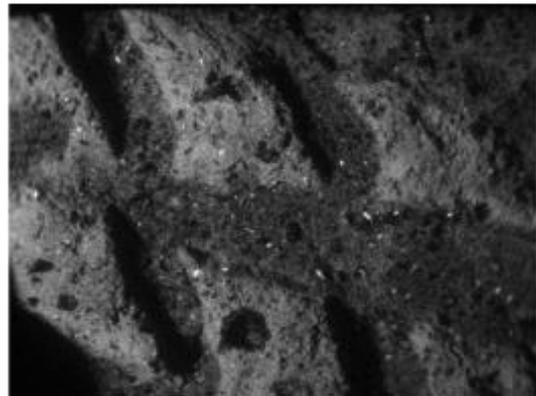**Figure 73:** Electron image of the area analysed with EDS (G1021)

Electron Image 13

Figure 74: Electron image of the area analysed with EDS (G1030)

B Data from Analysis No. 2

Ca K series

1mm

Si K series

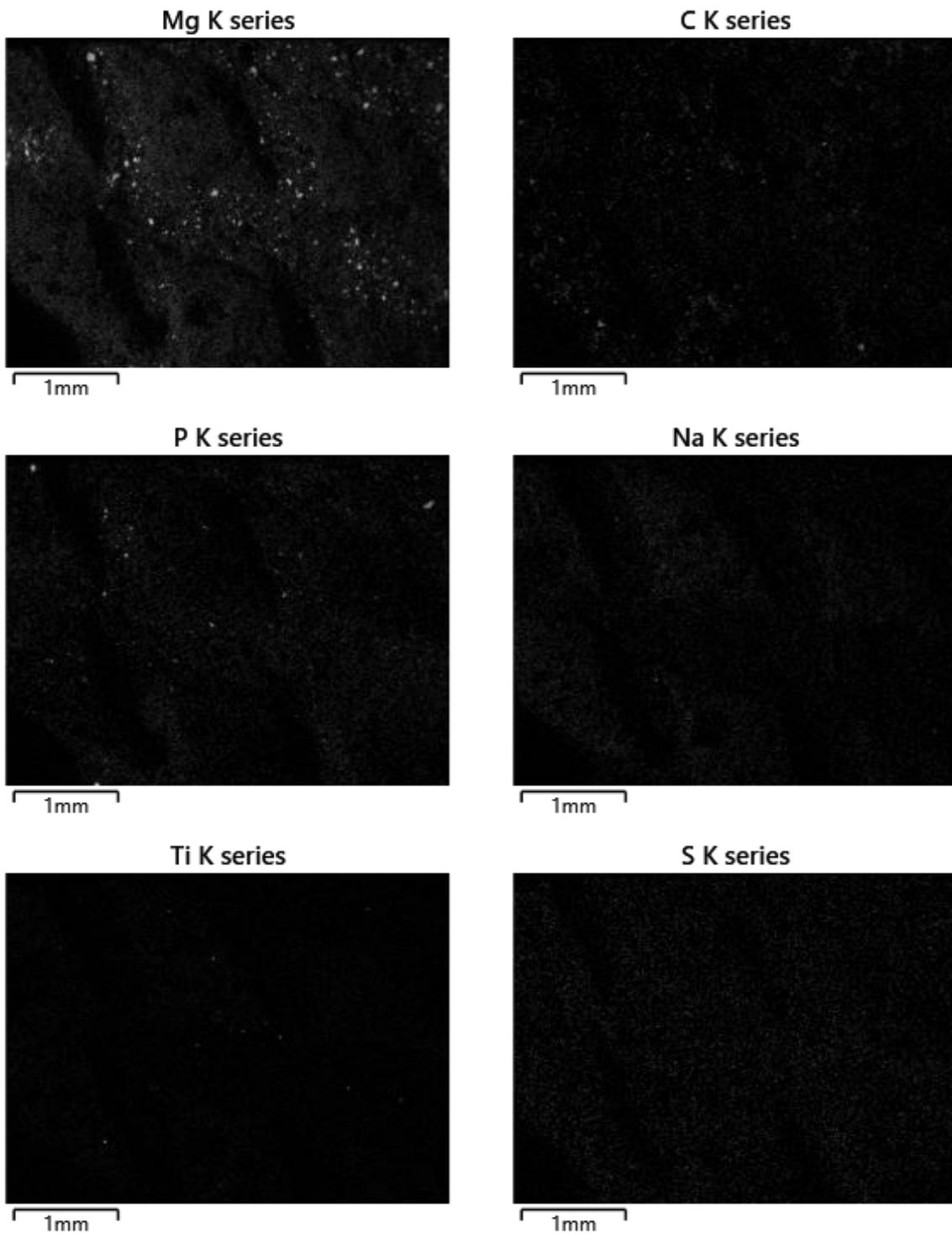1mm

O K series

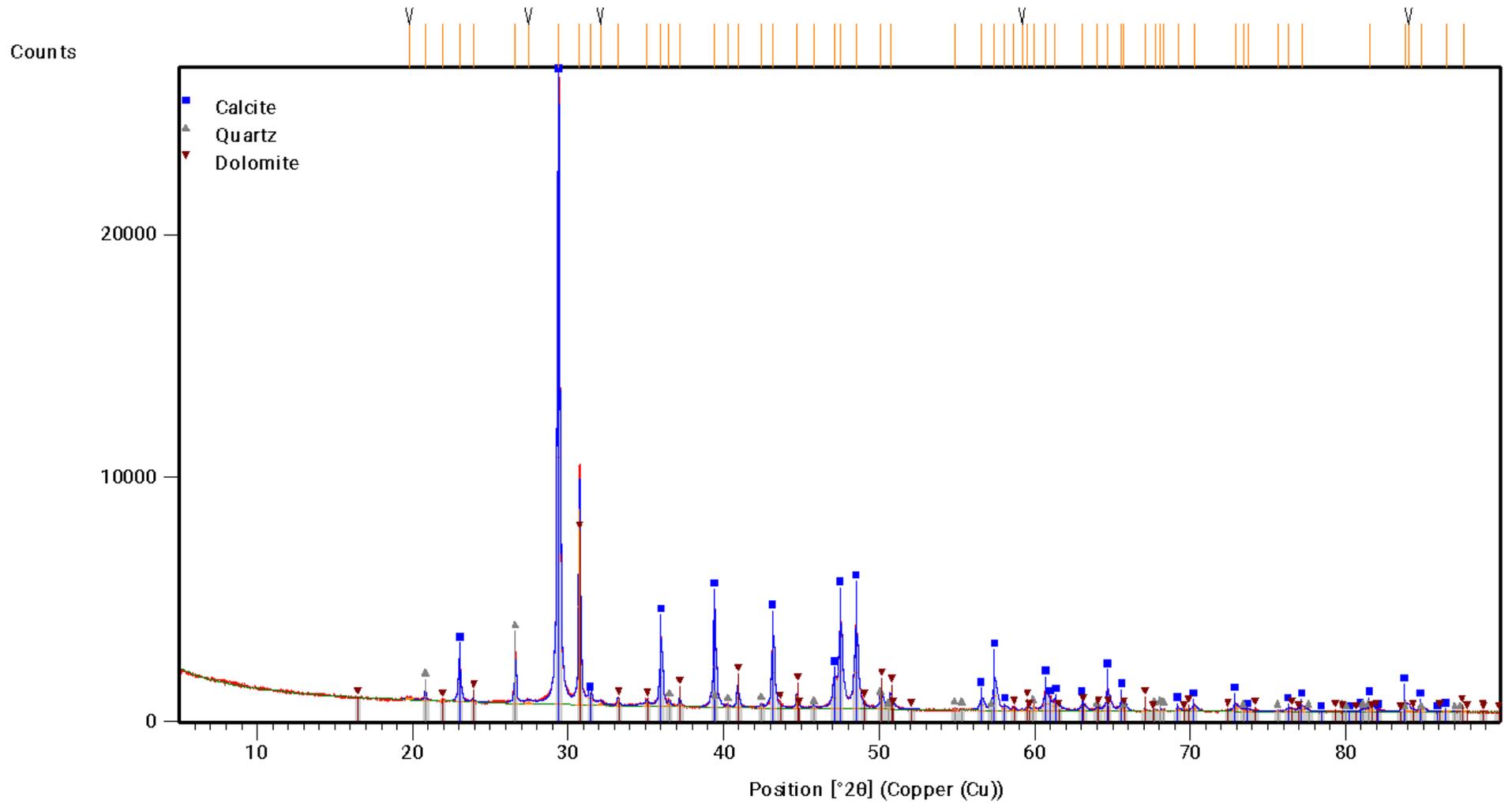1mm

Al K series

1mm

K K series

1mm
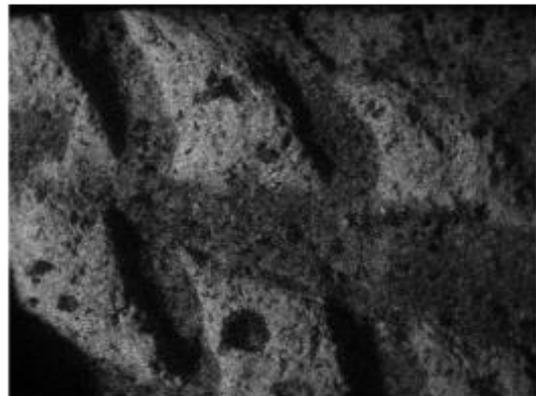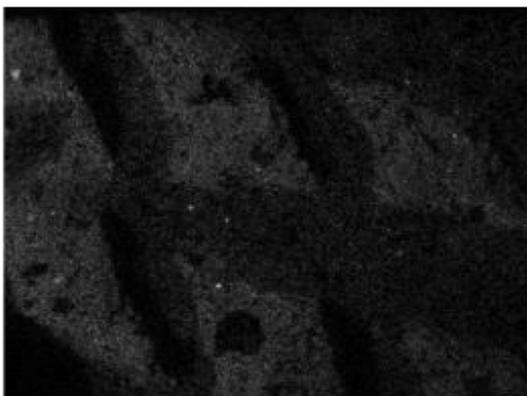
Fe K series
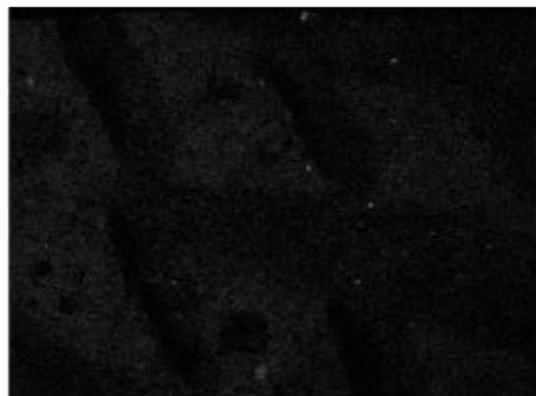
1mm

Figure 75: EDS Data of G1004 (analysis no. 2)

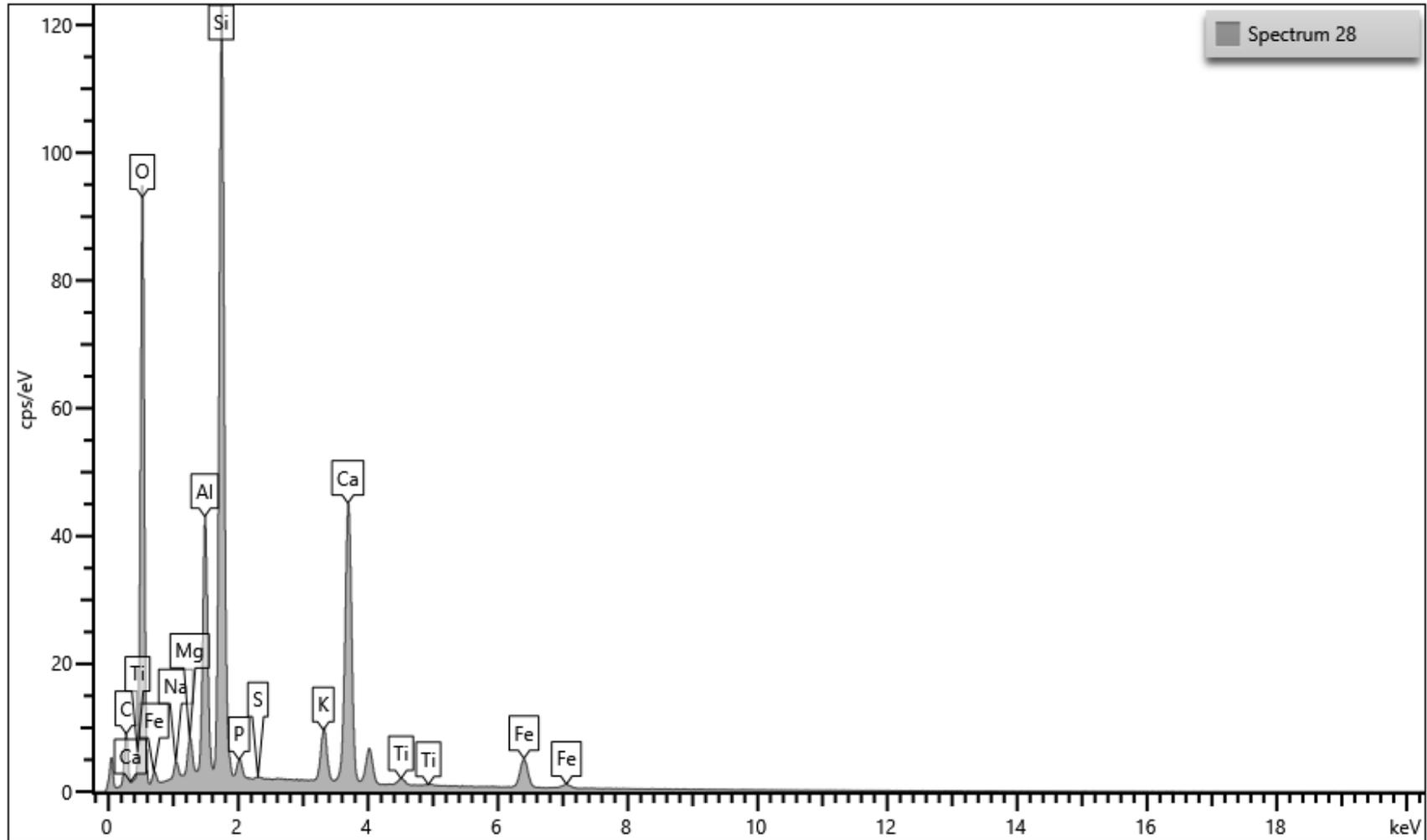**Figure 76:** Micro-XRD data of G1004 (analysis no. 2)

C Other Data

Figure 77: EDS data of the ceramic body area analysed for G1030