



Article

Recycling and Reuse of Building Materials in a Historical Landscape—Viminacium Natural Brick (Serbia)

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Abstract: During the MoDeCo2000 scientific and research project on mortars used in the territory of the Roman Danube Limes in Serbia, the biggest challenge was the quest for the provenance of raw materials. The area where Viminacium, the largest city in the province of Moesia Superior developed, with millennial continuity of land use and settlement, was selected as research case study and is presented in this research. People throughout history have always used what they had at hand, and the building remains were not only reused but also recycled for new constructions. Thus, the building material of Roman Viminacium has survived in the landscape through the in situ preserved remains of Roman buildings, as well as in the structures from the later periods, up to today. To the best of our knowledge, the use of natural sediments baked during the selfcombustion or combustion of underneath layers (coal in our case) for the purpose of construction was extremely rare in the Roman Empire. In this study, we follow the presence of this type of material precisely in Viminacium construction, naming it natural brick, while focusing on its potential use in lime mortars whose production was perfected in the Roman period and has never been surpassed afterward. Archaeological contexts in which this material was found have been studied, along with simultaneous work in the laboratory and in the field during the research and experimental use of the natural brick in lime mortars. We sought to determine whether this material could have been recognised by Romans in Viminacium as a potential valuable pozzolanic component of mortar, along with or instead of fired brick, being locally available and recyclable. The final confirmation of its pozzolanic features and later discussion open completely new directions for the future research of Viminacium lime mortars.

Keywords: Viminacium; natural brick; Roman mortar; pozzolan; natural clinker; coal combustion; building material; recycling; reuse; sustainability



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1. Introduction

Construction of various buildings was exceptionally developed during the Roman period in all territories that the Roman army reached. It was guided by the architecture developed in central Italy [1–4], but was blended with the traditions, knowledge, and experience of the local people that the Roman army conquered, and was influenced by the skills and background of soldiers, tradesman, and people of different origins traveling all around the Empire [5–7]. The Roman frontier, stretching for over 7500 km through Europe, Asia, and Africa [8], was the place where the contact, exchange, and communication between different people shaped all life aspects. It was the area where the Romans and barbarians met [9], where Roman culture was transmitted and where it also absorbed influences from the outside [10]. One of the sections of the frontier was the Danube Limes, 2000 km long, connecting today's Eining in Germany and the Danube mouth into the Black Sea [11].

The largest number of known or researched archaeological sites on the territory of today's Serbia, which originate from the Roman period, are located along the Danube river,

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once belonging to the Danube Limes. The only two legionary fortresses in the territory of the province of Moesia Superior, later Moesia Prima, whose borders broadly correspond to most of modern central Serbia, as well as parts of North Macedonia and Bulgaria, were Viminacium (Kostolac) and Singidunum (Belgrade), both located on the Danube Limes [12]. Beside the legionary fortresses, many auxiliary forts and observation posts were erected along the Danube [13], while major provincial cities developed as political, economic, and cultural centres [12]. The initial functions of civilian settlements by the fortresses followed the needs of the army, but over time they developed their own life, during which various activities took place, including construction.

The territory of the Danube Limes in Serbia has been chosen for the research of the project Mortar Design for Conservation-Danube Roman Frontier 2000 Years After (MoDeCo2000) [14], whose focus is on building materials, specifically lime mortar, as a binding material whose use and preparation from the Roman period has never been surpassed afterward, and simultaneously on the design of conservation mixtures compatible to historical mortars, using local raw materials. During the project research, the area related to the legionary fortress and the city capital of Moesia Superior, Viminacium, was singled out as an exceptional landscape example in which all aspects of civilian and military life on the Danube Limes during Roman domination, i.e., in the period from the 1st to the 6th century AD [15,16], could be followed, including building activities and exploitation of the natural resources. In addition to military structures, which include the ramparts, towers and gates of the fortress, and principium, large city facilities, such as amphitheatre and public baths were excavated, as well as peripheral villas, roads, aqueducts, and more than 14,000 graves and tombs [16]. There are many publications concerning everyday life in Roman Viminacium [15,16], but still very few on its building activities. However, the research into exploitation of raw materials, their trade, transport, and later use in the buildings, as well as construction processes conducted during the erection and later life of buildings that once existed in Viminacium, can offer an almost complete picture of construction on the Danube Limes in general.

The area of Viminacium is rich in coal, which is the reason it has been one of the most important industrial areas of Serbia since the 19th century. After decades of underground coal mining, strip mining has been developing here since the 1940s, accompanied by the production of electricity in thermal power plants [17–20]. Thus, the presence of this mineral resource has been shaping the life of the inhabitants and the natural environment for more than 150 years. However, the existence of coal was very important for the Romans as well, offering them baked sediment that they turned into a unique building material: the red material created out of the clayish sediments baked during the combustion of the coal layers beneath. We call this material natural brick.

The general context of this study is connected to the recycle and reuse of building materials in the ancient period. The course of economic activities related to the life of any product usually includes extraction, production, consumption, and disposal, but we must also add accompanying processes such as reuse, maintenance (including repair), and recycling to the list [21]. The processes of reuse and recycling are both related to the repeated use of some material, in its initial or new shape, but they are not always easily recognisable and mutually divisible while searching through the archaeological traces. Their nature is connected to the changes in the states of materials, which can be sometimes hidden from the observers and are the least visible in the case of the domestic everyday activities [21]. During recycling, materials acquire new forms and can also acquire new functions [22], when they are dismantled into constituent elements or even melted, and each activity requires energy for the incorporated processes [21]. In the process of reuse, no initial energy is usually needed to change the object, but on the other hand, cognitive processes related to its new meaning are included when the repurpose as a special type of reuse is conducted [21] and the materials obtain new functions. As an intermediate state between reuse and recycling, the researchers mention reworking, in which already used elements are further processed, but their overall state is not changed [22]. When we

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speak about the life cycle of a building in the contemporary world [23], it includes design, production, construction, use, and end of life, and the possible processes after its life ends that include maintenance (repair, renovation, any improvement), refurbishment (restoring to former better condition with alterations), demolition (disassembling or destroying), and deconstruction (selective dismantling for the purpose of reuse, recycling or repurpose) [24].

The term sustainability has been described with several thousand definitions among researchers [25]. One of the wide definitions determines it as "the balanced integration of economic performance, social inclusiveness, and environmental resilience, to the benefit of current and future generations" [25] (p. 766), and the other one states that it is "an indefinite perpetuation of all life forms" [26] (as cited in [25] (p. 758)). Circular economy as "a condition for sustainability," "represents a regenerative system in which resource input and waste, emission, and energy leakage are minimised by slowing, closing, and narrowing material and energy loops" and is achieved precisely through "long-lasting design, maintenance, repair, reuse, remanufacturing, refurbishing, and recycling" [25] (p. 767). It seems that both terms were embodied in the processes of the construction industry in antiquity.

After their use ceased, some Roman buildings were systematically deconstructed in order to obtain material that was later reused in the same locality or was transported further [22]. According to Roman laws on demolition, this process was not forbidden but legally regulated [27]. The demolition, salvage, and reuse, with recycling as the "routine" and a part of previous processes were "regular parts of the Roman building industry" [27] (p. 834). However, the research into the reuse and recycling of materials in Roman construction has been mostly conducted about marble and stone spolia [28,29], but still very little about other materials [22]. Spolia were used throughout the entire duration of the Roman Empire [30]. For example, in the 2nd century AD, the capitals of the Old Forum at Lepcis Magna were dismantled and used again in a porch between its amphitheatre and circus [31] (as cited in [27]). Their widespread use began after the crisis in the 3rd century, mainly from economic reasons, such as the lack of materials, artisans, and artists [30]. However, the most analysed examples are the reliefs taken from the triumphal monuments of Trajan, Hadrian, and Marcus Aurelius and incorporated into Constantine's triumphal arch in Rome, which are interpreted as the bearers of political message with Constantine as the successor of the great Roman emperors [30]. Finally, the abandonment of the temples during the Christianization with changes in city administration and planning created the real conditions for the secondary use of materials [30]. With a law from the year 397, it was allowed to use material from temples for bridges, aqueducts, and walls, and according to another law, the stone material from tombs was allowed to be taken for masonry of walls, as well as for decoration of halls or porticoes [30]. Additionally, the reuse was very often during the repairs after damages, such as earthquakes [27]. Regulation of the large-scale demolitions is visible from the example of calculations done by the researchers [32] (as cited in [27]). In Rome, 54% of imperial projects conducted from 180 to 305 AD were actually restorations; this number was 59% in the northern provinces and 24% in Asia [33–35] (as cited in [27]).

In the field of construction and decoration, Romans recycled a variety of elements made from stone, clay, wood, metal, or textile, among which are stone blocks or slabs, bricks, and mortars [36]. During the excavations of Viminacium, the reuse, repurposing, and reworking of architectural elements and tombstones, as well as building materials such as stone blocks or bricks, were detected in many ancient buildings and graves. In the recent excavations, a tombstone used as the cover for a sarcophagus was found, a lion sculpture built in a masonry structure of a modest building, as well as stone fragments with inscriptions, carvings, and holes, which were remodelled as stone blocks and built into the massive wall that bridged the ditch connecting the fortress to the city rampart [15,37–46] (Figure 1). Additionally, it is known that the remains of Viminacium buildings were demolished for the removal of building material in later periods and used for the construction or decoration of medieval fortifications, monasteries, and recent local houses, in the landscape itself but

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also in the surrounding areas. The monumental medieval Smederevo fortress is thought to be built of material coming from Viminacium, with visible *spolia* used as well. Embedded stone blocks, as well as Roman *spolia*, are visible in the walls of monasteries in the near vicinity of Viminacium. Finally, many local village houses were built using Roman bricks during the last centuries, and some of them are still standing today [46–55] (Figure 1).



Figure 1. Stone blocks in the massive wall in Viminacium (**top left**); *spolia* assumed to be from Viminacium built in the tower of Smederevo fortress (**top right**); *spolia* assumed to be from Viminacium built in the wall of the church in Nimnik monastery (**bottom left**); local house wall in the village of Kostolac built of Viminacium bricks (**bottom right**). Photo documentation of the MoDeCo2000 project.

Building materials used in Roman construction were almost exclusively obtained from local sources [56]. They were only transported over long distances because of rarity or because they were suitable and needed for some specific function, that is, mostly connected to large imperial and public buildings [56]. This does not only refer to stone, although it can be assumed that way while looking at the remains of once luxurious and monumental buildings, but to all materials, even the mortar.

Mortar is one of the most complex building materials in historical constructions, which was created throughout the millennia using aggregate and binder, that is, sand and rocks, earth, gypsum, lime, and cement, adding different additives and admixtures of the inorganic or organic nature to enhance its properties [57]. Among them are the natural or artificial materials with pozzolanic features that we call today the pozzolans. The pozzolan is a siliceous and/or aluminous material named after the ash from the Campi Flegrei volcanic district surrounding the Gulf of Pozzuoli (ancient Puteoli), near present-day Naples, that reacts with lime or compounds based on lime in the presence of moisture at ordinary temperatures, producing compounds with cementitious features [58] (as cited in [59]). The Latin name of the authentic material, also named pozzolana, is *puteolanus pulvis* ("dust of Puteoli") [59].

Roman lime mortar has been a debating point in the archaeological, geological, and material sciences communities for a long time. Although natural (mostly of volcanic origin) as well as artificial pozzolanic materials (mostly crushed ceramics–terracotta) were used before the Romans in the Mediterranean world, it was used for waterproofing mortars, that is, renders and plasters [60,61]. Only the development of hydraulic mortars, strong and water resistant, with the ability to set and harden under water [59,60], with the abundant use of natural pozzolanic materials of volcanic origin, enabled the full use of their potential for structural purposes, which resulted in the formation of so-called

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Roman concrete [59], composed of *caementa* (fragments of bricks, stones, and volcanic tuffs as aggregate) bonded by *materia* (volcanic ash mortar) [59,62], and the erection of the most well-known monumental Roman constructions. Volcanic ash is the product of pyroclastic eruption and is composed of glass and crystals originating from molten rock and particles of broken lava [59]. Tuff is the rock created from ash and lapilli, by lithification and consolidation, developing mineral cements, or sometimes through volcanic glass welding [59]. The pozzolanic mortar with volcanic material was thus a key component creditable for the durability of Roman constructions [62].

The ingredients for Roman lime mortars had to be obtained by exploiting natural raw materials that were not always readily available or in sufficient quantity and quality. Although their choice was closely related to the mortar function, it was also mostly guided by the economy. Thus, the local geological setting, the availability of the energy source for producing lime, as well as spatial relationship of the source of raw materials to the building site, were the most important factors in the activities in the production of mortars [63]. Many examples of mortars found in the structures around the former Roman Empire were produced using local materials that were not always of the best quality, but were "the best choice from an economic point of view" [64] (p. 145). At the same time, we have examples of the intentional mixed use of imported and local raw materials, but even in those cases, it was always economically justified as much as it was possible. It has been already established that Romans transported the natural volcanic pozzolanic materials, pozzolana, for mortars from the Campi Flegrei along the Italian coast, and it was also assumed that it was the case with the wide Mediterranean area [59]. Recently, during the research on mortars used in the harbor of Sebastos in Caesarea Maritima, Israel, volcanic pozzolanic materials imported from the Campi Flegrei were detected, but their use was "accurate," limited to the concrete-like structures and masonry walls in the intertidal zone, which confirmed the Roman optimisation in the supply of raw materials [65] (p. 19).

The research in this paper is related to one aspect of the construction activities in the Viminacium landscape, that is, the exploitation and use of local raw material for building purposes, with the assumptions made about its possible use in mortars as natural material with pozzolanic features. Through the overview of the red material created out of the clayish sediments baked during the combustion of the coal layers beneath, we will try to show an example of the historical sustainable use of building resources in the landscape. The focus will be given to the Roman period which left the largest number of material remains. Viminacium natural brick [46,53,66–68], more precisely naturally fired brick [69], is called "crvenka (reddish)" by the local community. Its bed is situated in the territory of the surrounding town and village of the same name (Kostolac) along with the former underground lignite mine, and it has been exploited for construction purposes from antiquity to the modern age [46,53,66–68].

Very noticeable ceramic-like reddish fragments, and the often-red dust that finally gave the colour to the mortar mixture itself, have generally been recognised in Roman mortars as originating from terracotta (fired brick, tiles, and amphoras or other pottery), thus telling us it was recycled from an already used material [70]. Vitruvius himself wrote about the use of crushed and ground brick in the flooring mortars and plasters applied in moist places [71] (VII. 1), and its presence has been historically attested in those mortars that needed resistance to water and moisture. The Viminacium baths represent a place where this important feature of the terracotta mortars, in Italian called cocciopesto [60], was very much needed.

The production of bricks and pottery was developed along the entire Roman Danube Limes, with Viminacium as a provincial centre, which has been attested by many brick and pottery kilns and manufacturing sites excavated in Viminacium. Its products were found during the research of many sites along the Danube, with pottery found in other provinces as well [72–74]. The production is certainly the consequence of the geological characteristics of the area in which Viminacium is located, which is characterised by the existence of loess, sands, alevrites, pebbles, clays, and coal [75], and which provided the inhabitants of this

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area with an abundance of brick raw materials not only in antiquity but also in later periods. The oldest found kiln in Viminacium was for brick and pottery, and dated to the end of the 1st century. A large brick and pottery production site was dated to the 2nd and 3rd centuries, the periods of the most intense brick and pottery production in Viminacium, while other excavated brick kilns in the wide Viminacium area are dated to the period from the 3rd to the 4th century [72,74,76,77]. Until the second half of the 20th century, there were numerous village family brick manufacturers, and with the development of industry in the 19th century, numerous brick factories were also established in the wider area [46,66].

During the MoDeCo2000 project, Viminacium mortars with an abundance of visible reddish fragments or simply being of a red colour, and almost exclusively used for plastering, rendering, and flooring, were sampled. Those having the mentioned functions originated from different structures dated to the period from the 2nd to the 4th century. Large reddish fragments, but in lesser amounts, were visible in a strong mortar for a sarcophagus's brick-built base, as well as in a bedding mortar of a villa, both dated to the 3rd century. Additionally, a sample of bedding mortar containing large fragments and originating from the 6th century early Byzantine rampart was researched (Figure 2). Were these fragments that we encounter in Viminacium mortars always created by crushing and grinding the bricks, tiles, or pottery, as usual, or were the origins of some of them connected to natural brick formation? The research on this topic started a few years ago [67], but in the recent period more research was mutually connected, mixing the archaeological view on the topic with laboratory investigations in natural sciences, offering initial chemical, physical, and mechanical characteristics of natural brick, and resulting in the creation and application of experimental mixtures of mortar using this material as an addition or admixture [68]. This research process provided us with important data that can be used for further research in the sciences, but also for future social and economic interpretations of activities of reuse and recycling related to building materials and construction in general in the Viminacium historical landscape.



Figure 2. "Brick" mortars from Viminacium (dating from the 2nd to the 6th century): (a) principium floor; (b) city baths floor; (c) city baths plaster; (d) city baths floor; (e) city baths plaster; (f) city baths plaster; (g) city baths plaster; (h) tomb plaster; (i) sarcophagus base; (j) villa floor; (k) villa bedding mortar; (l) early Byzantine rampart bedding mortar. Photo documentation of the MoDeCo2000 project.

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In this study, we will try to offer a review of the use of natural brick in ancient Viminacium constructions while referring to the archaeological traces. The study will offer observations on the possible use of natural brick in Viminacium lime mortars in addition to or as a replacement for the man-made fired terracotta products, using initial laboratory research to mutually compare different samples of natural brick, red tesserae in a sample of mosaic, and red fragments in the mortars of Viminacium, thus analysing if a natural brick has pozzolanic features, allowing it to be used for obtaining hydraulic lime mortars. In the discussion about the possible use of natural brick in mortars, its reuse and recycling are recognised as important topics.

2. Background

2.1. Historical Use of Natural Brick

The investigation of natural brick during the MoDeCo2000 project has been initiated by the assumptions made after long-term archaeological research of Roman Viminacium. Given that the past researchers of Viminacium encountered this material during excavations as fragmented materials in wall cores and road foundations, its role was not considered as important for drawing conclusions about construction in this city and legionary fortress. However, through analyses of the excavated remains of Viminacium masonry structures, among which were the remains of the northern gate of the legionary fortress partially researched in 2022–2003 [16] with the rubble of natural brick in the foundation level, and their comparison with research notes from the beginning of the 20th century in which the periods of Viminacium life were connected with the use and combination of different building materials (wattle and daub, fired brick with schist, and rubble) [78,79], the use of natural brick was connected with Viminacium's earliest building phase dated to the 1st century AD [53]. After the beginning of large-scale systematic archaeological excavations of the legionary fortress of Viminacium in 2016, it was more precisely dated to the period of the last decades of the 1st century AD [43], which was the result of the first extensive Roman building activities in the area; the assumption that natural brick was predominantly used during this phase as the basic building material was proven.

During the Roman period, the province of Moesia Superior was a mining area with rich mineral resources: gold, silver, lead, iron, and copper [80]. However, Viminacium's narrow territory, as a part of the Kostolac lignite basin, was rich in coal instead [46,81]. Its exploitation was not mentioned in sources until the 19th century, and underground mining officially started in 1873 [19,20,46,67]. Thus, it is not known if Romans of Viminacium exploited coal as a fuel, but it is possible that they knew about its presence, since its layers were close to the surface [18,67]. It is written by a chronicler of the Kostolac underground mining that the coal layers were revealed from the surface and thus enabled simultaneous coal exploitation and exploration works [18]. In 1875 the coal spontaneously combusted, and in 1890 it was noted that it cannot be excluded that the coal was known here for a long time, although the first records are dated after 1870 [18].

Raw wood and raw wood made into charcoal were the most-used fuels by the ancient people, while the non-wood fuels and coal were used to a much lesser extent [82]. Most of the coal finds in Roman Britain are connected to metallurgy [83] (as cited in [84]). According to the coal finds in the hypocaust of the baths [83] (as cited in [84]) and their distance to the coal seams, the researchers concluded that coal was transported within the coast [85] (as cited in [84]). From the 1st century AD it was used for domestic purposes and metallurgy; its maximised use was dated to the period from the 2nd to the 4th century, and it remained in wide use in the 5th century, after which it was not used for centuries [86–88]. Along with the predominant use and presence of wood, it seems that it was not economically feasible to widely transport the coal, or maybe the available coal resources were not rich enough for long-term exploitation [84]. Research in Britain also showed that the Romans did not mine the coal from the deep, but exploited it from the deposits on surface [82], that is from the exposed seams, using simple tools [83] (as cited in [84]). Thus, since the coal in Viminacium area was locally available, and probably accessible from the surface, it is very

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much possible that the Romans exploited it and used as a fuel, but it is less possible that they mined it from the deep extensively, since the wood was abundant in the area until the 19th century and the development of the mining industry [18]. In the composition of many examples of historical mortars, the remains of charcoal are visible. The reason for their presence has still not been precisely agreed upon among the researchers. Since, their amount is generally very small so as to be considered aggregates, help mortar to "breath," or to contribute to the colour, they were probably the remains of the fuel used for the lime burning, thus present as impurities [89]. During the research of Viminacium mortars in the MoDeCo2000 project (26 samples), the charcoal inclusions were not observed.

One of the historical mining records, originating from 1875 and the time of the underground mining in Kostolac in the area of the crvenka bed, informs us about coal fires in the mine and a specific combustion material: "At the entrance to the underground... red clay with fossils is observed. This metamorphosis was caused by fire, which is a very common case with coal found in the soil. In other geological ages, coal may have been fired and burnt. This kind of combustion affects the surrounding rocks and changes them in various ways" [18] (p. 79). In this description, we recognise the natural brick.

Since its bed is located in close surroundings of Viminacium (Figure 3), natural brick was very available for its construction. Its high availability was surely considered the most important and favourable feature during the process of selecting materials for various construction needs by the first builders of Viminacium. The walls formed from hewn natural brick blocks were first recorded and explored during the systematic excavations in 2016, when it was detected that the ramparts and towers of the first fortress of the Legio VII Claudia from the last decades of the 1st century AD were built from this material, as well as the walls of the principium building, whose excavations started later (Figure 4). Additionally, channels in the fortress were built of these blocks as well, the wall foundations and cores were made of fragmented natural brick; the substructures and final layers of floors were created from crushed or almost ground natural brick, while its small fragments were also used in the formation of the embankment along the rampart [43,45,90–93] (Figure 4). Use of natural brick was developed before the locally produced fired bricks and schist, quarried in the nearby villages of Ram and Zatonje, became the main building materials of Viminacium, resulting in somewhat less use of limestone. In the near vicinity of Viminacium, there are no widely known sources of limestone. The source of the limestone blocks found in its buildings has been the topic of different studies over years, but the final conclusions have not been published yet [53].

Natural brick was also found in the foundations of walls and floors, as well as in overground wall structures of modest auxiliary facilities and buildings of peripheral rural estates during later periods, such as structures dated to the wide time span from the 1st to the 4th century [94], those connected to the period of the first half of the 3rd century [39,95], and in the buildings of unknown purpose dated to the periods of 3rd and 4th century [96]. In the area of the amphitheatre, as well as in the fortress area, we see it under the later phases made of stone, where the ruined structures made of natural brick from the 1st century were either levelled and used as foundations or substructures of the later wall and road structures, or were just incorporated into younger structures [42,43,45,90–93,97–99].

The known traces of the use of these types of combustion products in the Roman and later periods are generally very scarce. Natural brick from Kostolac was used during the construction of Roman Viminacium, but also in the buildings of another nearby Roman city, Margum (Dubravica), situated on the confluence of the river Morava to the Danube in present-day Serbia. In its earliest structure, dated to the 1st century,"pieces of loess of irregular shape baked on fire in the coal seam below it" from Kostolac were found [100] (p. 119). We do not have records of its use in the Viminacium area before the Roman conquest nor in the Middle Ages [50,101,102]. In England, the burnt mudrocks (shales) from the cliffs at Dorset Coast [103] were used for decorative flooring, that is, mosaic tesserae and slabs for opus sectile technique, in Silchester Roman town, Fishbourne Roman palace, Angmering Roman villa, and Eccles Roman villa, as well as in Caerleon Fortress

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baths in Wales, and were dated to the mid 1st and early 2nd centuries. It was assumed that the raw mudrocks from the beds in Dorset that were not naturally burnt were probably only collected here and later burnt on the beach or nearby, being transported afterward, while only a limited quantity of sediment burnt on site after self-combustion, due to their organic-rich nature, was additionally used [46,67,104–107].



Figure 3. Position of Viminacium and today's Belgrade (Singidunum) along the Danube in Serbia, and a close-up of the position of the exploited parts of the bed of natural brick. Tags: authors of the paper on Google Earth Pro photo printed on 18 December 2022—historical image from August 2022.



Figure 4. Natural brick in Viminacium legionary fortress, used in the wall of the principium in the form of blocks (**top left**), and the rampart with natural brick in the form fragments used in the foundations and wall core, as well as fragmented blocks used over ground, along which the later stone phase was erected (**bottom left**); different fragments of used natural brick collected from the principium site (**right**). Photo documentation of the MoDeCo2000 project.

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However, the archaeological traces of the use of these natural products by humans are numerous in prehistoric sites [108–111]. It is known that these ceramic-like rocks were used by the early Native Americans for tools and blades in Wyoming and Montana [111]. Natural clinkers from sub-arctic Canada were used by hunter-gatherer communities in North America for the same purpose from 10,000 years ago, until the arrival of Europeans [109]. They have been also found during archaeological excavations and detected as being used by humans for the polishing of tools during the prehistoric period in the Serbian Kolubara lignite basin [112].

It is interesting to mention here the so-called FCR (fired-cracked rocks), whose presence is attested by archaeological research worldwide. The beginning of use is dated to the period 32,000 years ago, and they were created by deliberate heating of rocks [113]. Their most common use was for cooking, where one of the methods was to cook food slowly in an earth oven, without fire but with previously heated stones. The other supposed use was for bathing, when heated stones were submerged in the pits with water for the release of steam [113,114]. Experiments showed that features of the rocks after their thermal alteration depended on whether they were cooled slowly or quickly [113]. The prehistoric examples researched in Ohio, USA, are analogous to the most present type of natural brick in the natural bed of Kostolac, and were created by firing the sandstone [114]. FRC was also used for flooring in metal-making furnaces, ceramic and charcoal kilns [113], heating of shelters, enclosing the hearths, etc [115], and could have been reused several times [116] (as cited in [114]), also functioning as different tools [115]. It is not always easy to distinguish the FCR from naturally fired rocks, and the use of different laboratory techniques to achieve this is needed [117,118] (as cited in [114]).

Natural brick in the area of Kostolac was exploited until recently for different building purposes, such as for the surfacing of industrial and village roads [46,66,67] from three areas of its bed whose total reserves in 2002 were estimated to be 1,205,000 m³ [119]. It is extensively used for roads in areas of the USA where it is abundant, and better-quality material is not readily available locally and is expensive for transportation. There, it is classified as one of the types of crushed stone [120,121], covering one-third of all crushed stone aggregates for the roads in Wyoming [121]. However, tests in New Mexico showed its variable and marginal quality for use in final layers, considering abrasion. Resurfacing in Arizona needs to be performed every 3–6 years depending on its type and uniformity. It has, however excellent drainage properties and is used in mines to control erosion and to stabilise slopes, as well as a base material below concrete slabs [120]. This material was also used in the mine of Santa Barbara, Cavriglia, Italy during the 20th century, for paving mining tracks [122]. Thus, the common functions of this material throughout the centuries in Kostolac and in the world are connected to the building of road infrastructure.

2.2. Geology of Natural Brick

The material consisting of different sedimentary rocks created by their burning or melting due to the ignition of coal in their immediate environment is found in many places in the world and is known by different names, depending on its type, among which are clinker, natural clinker, porcellanite, laterite, buchite, and paralava, but also pseudoscoria and scoria [69,108,109,122–135], along with the more general terms of vitrified or fused and burnt rock [136]. *Scoria* is used as a local name in some USA states, which is, however, incorrect. Namely, the old researchers did not recognise these rocks as metamorphic but thought they were volcanic, and thus named them scoria [110]. The process is called pyrometamorphism or combustion metamorphism [109]. The material itself is called natural brick [46,53,122] in this paper, since it is a brick-like natural product, and in the case of Roman Viminacium, it had the function of a building material formed in elements for the use in construction [43].

One of the combustible mineral raw materials in nature is coal, which can be ignited naturally, that is, by spontaneous combustion in contact with oxygen, after lightning, or by wildfires, but also due to man-made fires [123–125,135]. Coal beds can burn when

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they are relatively shallow, meaning they have been unearthed to depths less than a few tens of meters from the surface, which are adequately ventilated and above the water table [137]. The earliest evidence of this natural process is found in the USA (Wyoming and Montana), and began at least two million years ago, producing coal clinker [138]. Hundreds of natural, accidental, or deliberately initiated coal-seam fires are burning today around the world [138]. Due to coal mining, their number has increased over time in the world, resulting in an ecological catastrophe because of the huge quantities of gases and particles, land subsidence, products polluting water and soil, displacement of communities, human disease and death, and the destruction of natural habitats [139]. It is estimated that 75% of coal fires were spontaneous [140] and occurred in deposits during the recent geological past [125]. The fires burn until they permanently spend all available fuel (coal) and oxygen, which means that some can be extinguished quickly on their own, or can burn for decades, or centuries [141]. They can burn even for the longer periods. An Australian underground coal seam named "Burning Mountain" is estimated to have been burning for the at last 6000 years, according to some researchers [142], or even 500,000 years according to others [143]. Fires are mostly extinguishable by human interventions, but due to the costs, risks, scale of the fire and its underground position, and unpredictable nature, it is very hard and sometimes even impossible to accomplish it, and the fires can burn for an indefinite period of time [139].

To most coal geologists, clinker "refers to a rock sequence altered by an adjacent coal bed burning in place instead of the manufactured consequence of an industrial operation or electrical generation process" [120] (p. 188). It includes different thermally metamorphosed or melted rocks, depending on the source rock, dynamics of burning, ventilation, and proximity to the fire. They range from thermally altered but not melted rocks (dubbed burnt or baked rocks), partially fused rocks (clinkers), to totally melted rocks (paralava), with porcelanite being a specific type of clinker heated near the point of melting [109,144]. The rocks are characteristically multi-coloured [145]. In 1929, a similar division was made for the material present in the coal seam in the Most Basin of the Czech Republic [145]: bricklike rocks (clinker); baked but not sintered clays produced by low to moderate thermal alteration (pale red or yellow coloration); porcellanites, dense, partly or completely sintered clays of jasper- or porcelain-like appearance with conchoidal fractures (pale grey, yellow, apple green, or reddish colour); and ferruginous slags and paralavas formed by the fusion of various carbonates, more or less holocrystalline, with common drusy vugs (from greenish to black colour [146] (as cited in [145])). Thus, its characteristics range from an entirely or partially fused rock, when it is very close to a source of fire, to slightly baked rocks when it is created at lower temperatures [147]. Similar characteristics are possessed by the natural brick in the bed in the village of Kostolac (Figures 5 and 6).

In Serbia, material created after the coal combustion is encountered in basins of lignite, which is a low-rank coal with great tendency to self-heat because of its high moisture and oxygen content [119]. All Serbian formations originate from the Pontian age [75,148], belonging to Upper Miocene epoch [149]. In the area of the Kolubara coal basin, these baked clays are called brand [148]. An interesting example is found in the Czech Republic, where the Medlovický deposit of this material, proclaimed as natural monument, was created above a coal mine after clay burning, and the material name is very similar to the Kostolac name—"červenice" and "červenka" [46,150,151].

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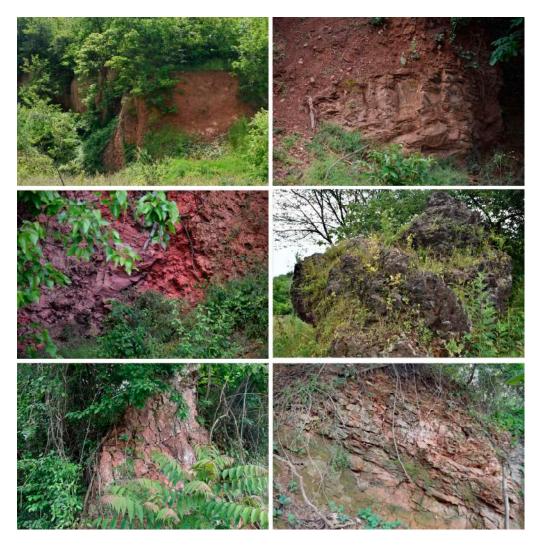


Figure 5. Different locations in the natural brick bed in Kostolac village with visible variety in colours. Photo documentation of the MoDeCo2000 project.



Figure 6. Cont.

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Figure 6. Different locations in the natural brick bed in Kostolac village with visible variety of colours, present as dark formations close to paralava, porcelanite and clinker, and baked clay. Photodocumentation of the MoDeCo2000 project.

3. Research Methodology

Many samples of natural brick for laboratory research in the project were taken from the bed (Figure 7), as well as obtained from ruins during archaeological excavations of the principium in Viminacium legionary fortress (Figure 4). Initial analyses, using macroscopic observations, were performed on samples of natural brick originating from one of the three bed areas that is closest to the remains of Viminacium; on tesserae, nucleus, and rudus of the accidental find of the mosaic fragment; and on the flooring mortars from the city baths and principium.



Figure 7. Various natural brick samples from the bed (**left** and **right**). Photo documentation of the MoDeCo2000 project.

Chemical analyses and tests on the physical and mechanical characteristics were carried out on two different bed samples (samples 1 and 2 in the further text) (Figure 8). A standard test for the determination of a material's pozzolanic activity was undertaken for these samples as well, later making an experimental lime mortar with this material as an admixture. The experiment was conducted during the workshop masonry work while building a small structure in Viminacium Archaeological Park.

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Figure 8. Two samples of natural brick from the bed, cut for the testing in laboratory. Sample 1 (**left**), sample 2 (**right**). Photo-documentation of the MoDeCo2000 project.

A mosaic floor fragment was found by a local villager in the field, and it is undated. It was visually examined, since an assumption was made that its tesserae were made of natural brick, as well as red fragments in its nucleus and rudus. Since the principium was built of blocks of natural brick, an assumption was made that red fragments in its floor were created by crushing and grinding the same material. Considering the mortars from the baths, they were used for visual comparison, since the red admixtures in their matrix were assumed to be fired brick, which was the material used for its erection, along with schist. The samples of various fired bricks from Viminacium were also used for a visual comparison with natural bricks.

During this research, the obtained results were compared with the already published results on Viminacium fired bricks [152], baked clays that were once used for plastering a Viminacium brick kiln [153], and with those of natural brick sampled from the same bed, but in a different area, previously published during the research of its potential use in the modern building industry [154] (sample 3 in the further text).

Scientific research mentions or confirms the pozzolanic features of this natural formation present in various forms around the world [155–158], which depend on the creation conditions, the firing temperature, and the primary rock. The previous research on the potential use of Kostolac crvenka in industry showed its suitability for protective mortars with excellent thermal and hydro potentials in agricultural and industrial buildings, as well for the production of decorative renders and plasters [119]. It was proposed that it should be used (as well as Kostolac loess, clay, gravel, and sand) in the industry of tiles, facade bricks, decorative ceramics, and concrete construction elements [159].

3.1. Macroscopic Observations, Methods and Results

A visual inspection of five natural brick samples from the bed was performed in order to define their colour according to the Munsell Rock Colour Chart, but also using a Dino-Lite USB Microscope (model AF4915ZTL) to obtain results regarding the samples' texture and presence of inclusions, in accordance with EN 12407: 2019 [160]. The samples from the bed were chosen according to their visible differences, from highly baked to those less thermally changed. Thus, they show obvious mutual differences in colour, shade, and texture, as well as in the presence and type of inclusions. These burnt sedimentary rocks are mostly highly fractured and cracked in the bed, and may occasionally contain faunal remains (Figures 7 and 8).

A mutual comparison of the fragments of natural brick sampled from the bed and the two mosaic tesserae showed their great similarity in colour and composition; the colours of two samples from the bed were determined as identical to the tesserae, according to the Munsell Rock Colour Chart (Figure 9). This can speak in favour of our assumption that the tesserae were made of natural brick, the type of which was, however, probably chosen to

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be resistant to wear since they were part of the surface layer of a floor. Red fragments in mosaic layers resemble one of the tesserae.

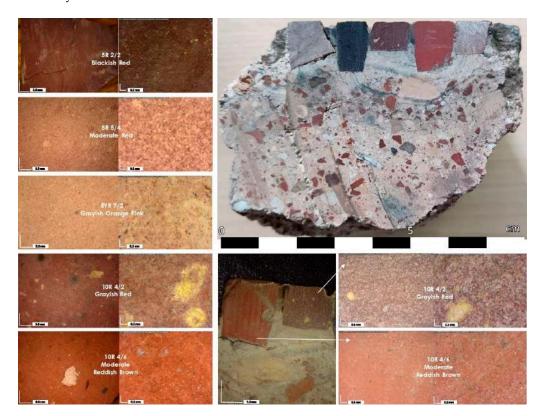


Figure 9. Colours of the natural brick samples from the bed (**left**) compared to those of mosaic tesserae (**right**). Photo documentation of the MoDeCo2000 project.

Floor layers made of crushed natural brick were detected in the first Viminacium structures connected to the fortress [43,45], which could mean that its builders also assumed or were already acquainted with the waterproofing and drainage features of natural brick. Although many mortars initially characterised as brick mortars were sampled from Viminacium buildings, a mortar sample taken from the floor of the central building of the legionary fortress, principium, and dated to the 2nd century, was assumed to have natural brick in its composition. It has the largest amount of ground "brick" used for the formation of its binder matrix compared to all mortars sampled during the MoDeCo2000 project from Viminacium. A visual comparison between cross-sections of a mortar sample used for flooring in the Viminacium baths, dated to the period of the 4th century, mortar forming the nucleus and rudus in a mosaic fragment that was assumed to have red admixtures of natural brick since its tesserae greatly resemble the natural brick itself under the microscope, and floor mortar from the principium, was conducted using the microscope. It resulted in a high visual similarity between the first two, but did not offer reliable data on this topic in the case of the third sample (Figure 10).

At the same time, the samples of natural brick from the bed were initially visually compared to two fired brick samples of similar colour originating from different buildings, collected during the excavation, and thus dated to different periods (Figure 11). Most of these bricks formed the construction of graves that were disassembled after the excavations because they were endangered by the surrounding mining industry. These funerary structures are mostly dated to the 4th century, but since the bricks were often secondarily used in the graves, this date is not reliable for their dating. Visual similarity of natural and fired bricks actually presents one of the difficulties in the initial determination of red fragments in mortars originating from natural or fired brick, showing the need for laboratory analyses with reliable methods of determination.

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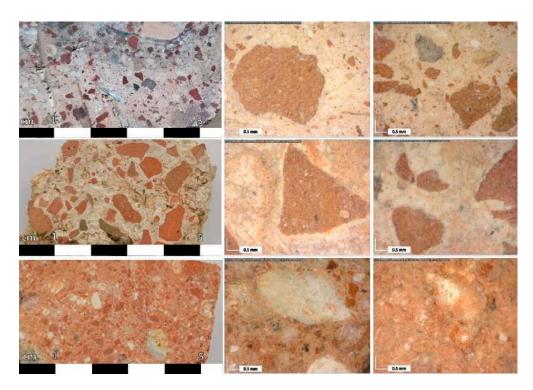


Figure 10. Red admixtures in the mosaic fragment of unknown date (**top**), flooring mortar from Viminacium city baths dated to the 4th CE (**middle**) and principium, dated to the 2nd century (**bottom**). Photo-documentation of the MoDeCo2000 project.

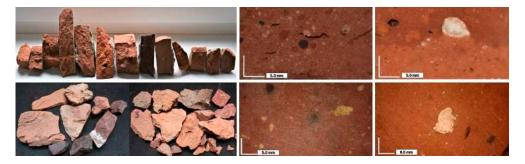


Figure 11. Fragments and colours of the fired brick samples (**top**) compared to those of natural brick samples from the bed (**bottom**) Photo documentation of the MoDeCo2000 project.

3.2. Physical and Mechanical Characteristics, Methods and Results

Compressive strength testing was performed using standard EN 1926: 2010 [161], in a hydraulic press, on cubes of natural brick obtained from two bed samples, named sample 1 and sample 2, chosen as being highly mutually different according to the visual observations. One of them was of a dark red colour which can be associated with the rock being baked at higher temperatures and sintered, as clinker or porcelanite, while the lighter one can be connected to less baked rock. The first one had cavities and the other had visible inclusions (Figures 8 and 12). The testing procedure was, however, modified since the dimensions and the number of cubes were limited to the dimensions of the bed samples. Thus, two cubes with the sides dimension of ~40 mm, per sample, were tested (Figure 12). The structure of the material could not be comprehended completely without cutting it into fragments. After cutting, it was visible that sample 2 was compact and did not have any direction preference in the structure, being similar to the fired brick, thus the specimens of this sample were cut as cubes. However, sample 1 had a great number of cavities. We supposed that its features would not be the same in all directions because of the present cavities, which themselves had a preferred direction. Since the specimens of

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sample 2 were cubic, we decided to unify the shapes but to pay attention to the direction of the compression force compared to the prevailing direction of the cavities in them. The cube marked as 1A was tested so that it was positioned with cavities predominantly spread in the same direction as the compression force, while the cavities of cube 1B were more spread perpendicular to the force direction. Their apparent density, water absorption, and absolute porosity were determined as well, according to the standards EN 1936: 2006 [162], and EN 13755: 2009 [163].



Figure 12. Test cubes of natural brick from the bed (**top**), and the position of cubes in the hydraulic press (**bottom**). Cubes (**1A**,**1B**) are cut out of the sample 1; Cubes (**2A**,**2B**) are cut out of the sample 2. Photo documentation of the MoDeCo2000 project.

Testing of physical and mechanical characteristics of the samples of natural brick from the bed named 1 and 2 (Table 1) showed that their cubes with yellow inclusions in the form of spots (2A and 2B), although with no cavities, had lower values of compression strength, namely 3.36 MPa and 1.42 MPa, than the cubes with no inclusions, but with spread cavities probably left after organic inclusions had been burned out (1A and 1B), which were 20.67 MPa and 4.33 MPa, respectively. It is visible during the testing of compression strength of the cubes 1A and 1B that extremely mutually different results were obtained.

| Table 1. Physical and mechanical | l characteristics of the samp | les of natural brick | from the bed (data |
|---|-------------------------------|----------------------|--------------------|
| for the sample 3 are taken from [3 | 155]). | | |

| Sample/Cube | Compression Strength (MPa) | Apparent Density (g/cm ³) | Absolute Porosity (%) | Water Absorption (%) |
|-------------|-------------------------------|--|--------------------------|-------------------------|
| 1A | 20.67 | 1.547 | 41.73 | 23.67 |
| 1B | 4.33 | 1.485 | 44.07 | 30.05 |
| 2A | 3.36 | 1.682 | 36.65 | 22.06 |
| 2B | 1.42 | 1.966 | 25.95 | 12.76 |
| 3 | 1.98 | 1.827 | 28.07 | 15.48 |
| | | | | |

The compression strength tests on seven *Viminacium* fired bricks dated from the period of the 3rd (one sample), 4th (five samples), and 6th century (one sample) were previously carried out and published [152]. The values varied from 9.73 MPa (4th CE) to 21.79 MPa (6th CE), with an average value of 17.67 MPa. The compression strength of the natural brick cube 1A broadly corresponds to the values measured for two out of seven fired bricks (4th and 6th century). The average apparent density value for fired bricks was 1.78 g/cm³, which is closest to the natural brick sample taken from the other area of the same bed

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during the other research, sample 3, which had a compression strength in the range of our sample 2, and whose absolute porosity and water absorption are closest to cube 2B. Thus, since the previously published research did not include photos of sample 3, we can only initially assume it was more similar to our sample 2. One of the tested fired bricks, whose research was published, was later chemically analysed. Its values of compression strength and density were 17.26 MPa and 1.70 g/cm^3 [152].

3.3. Chemical Analyses, Methods and Results

The chemical composition of two presented samples from the bed was determined using an X-ray fluorescence examination. First, they were dried at 105 °C until they reached a constant mass. Using an approximately 5 g powdered clay sample and 1 g of tableting wax (Cereox wax, Fluxana) per sample, the samples were formed into pressed pellets (diameter 40 mm) with a laboratory hydraulic press (Specac) applying a force of 20 t. The Energy Dispersive X-Ray Fluorescence (EDXRF) equipment, Spectro Xepos, was used in conjunction with a binary cobalt/palladium alloy thick target anode X-ray tube (50 W/60 kV) and combined polarised and direct excitation to perform the analyses. The detector for the Spectro Xepos was a silicon drift detector (SDD) design with a Peltier cooler device. The Spectro XRF Analyzer Pro Xepos C software managed the EDXRF analysis. For this investigation, the method of fundamental parameters for oxide testing was used.

The results of the chemical analysis for two bed samples (Table 2) show that the values of the sum of silica, ferric oxide and alumina, that is $SiO_2 + Fe_2O_3 + Al_2O_3$, for the samples 1 and 2 are 89.52% and 86.48%, respectively, fulfilling the chemical requirement for natural pozzolanic materials according to the ASTM C 618-22 standard [164], having this value higher than 70%. In addition, the sulphur trioxide (SO₃) content is less than 4.0% for both samples, while the loss on ignition at 1000 °C is less than 10%, which also complies with the same standard. As for the samples of baked clay from the Viminacium brick kiln, the sum of the mentioned oxides ranges from 73.73% to 92.75% [153], while the sum in the analysed Viminacium brick dated to the 4th century is 85.1% [152]. The SO₃ content and loss on firing at 1000 °C in bricks and baked clays also meet the mentioned standard. The mentioned sum of the oxides in the fired brick is slightly lower than in one of the samples of natural brick, while the highest value was obtained for a baked clay. The highest values of ferric oxide, alumina, and calcium oxide, CaO, are recorded with clays as well, while the greatest value of silica was detected for a fired brick. The contents of silica and calcium oxide are higher in the fired brick than in natural bricks, while the reverse situation is visible with alumina and ferric oxide.

Since scientific research on natural pozzolanic materials often shows contradictions between their real performance and fulfilling of the given standards, it is advisable to conduct additional research according to additional standards, as well as additional chemical, mineralogical, and thermal analyses [165]. In this study, a test for pozzolanic activity for both samples of natural brick was conducted.

Table 2. Chemical composition of natural brick samples from the bed compared to baked clays used for the plastering of a Viminacium brick kiln and a fired Viminacium brick (data for baked clays from the brick kiln are taken from [153,166]; data for fired brick are taken from [152]).

| Sample | Loss on Ignition (1000 °C) | SiO ₂ | Al ₂ O ₃ | Fe ₂ O ₃ | CaO | MgO | Na ₂ O | K ₂ O | SO ₃ | P_2O_5 | MnO | TiO ₂ | Total |
|---------------------------------|-------------------------------|------------------|--------------------------------|--------------------------------|---------------|---------------|-------------------|------------------|-----------------|---------------|---------------|------------------|------------------|
| Natural brick (1) | 1.23 | 61.97 | 19.36 | 8.19 | 1.70 | 3.01 | 0.46 | 2.18 | 0.06 | 0.17 | 0.28 | 0.97 | 99.57 |
| Natural brick (2) | 2.29 | 61.44 | 17.88 | 7.16 | 3.71 | 3.09 | 0.87 | 2.06 | 0.05 | 0.17 | 0.24 | 0.89 | 99.86 |
| Baked clays from the brick kiln | 1.56–9.27 | 51.26- 68.21 | 15.01- 21.88 | 2.66- 8.53 | 1.03- 8.36 | 0.73– 4.70 | 0.71- 1.38 | 1.54– 2.94 | | 0.05- 0.86 | 0.02- 0.16 | | 99.96– 100.34 |
| Fired brick | 4.09 | 69.40 | 10.50 | 5.20 | 4.42 | 2.95 | 1.59 | 1.76 | | | | | 99.91 |

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3.4. Pozzolanic Activity, Methods and Results

The preparation of mortar mixtures for testing pozzolanic activity of natural brick was conducted according to SRPS B.C1.018:2015 [167], while the test on mechanical features was carried out according to EN 196-1:2017 [168]. Hydrated lime was mixed with ground natural brick, sand (according to CEN standard), and water in the ratio of 1:2:9:2, that is, $150~\rm g \pm 1~\rm g$ standard hydrated lime; $300~\rm g \pm 1~\rm g$ natural brick; $1350~\rm g \pm 5~\rm g$ sand (CEN standard sand); and $300~\rm cm^3 \pm 1~\rm cm^3$ deionized water. Ground natural brick was obtained from samples 1 and 2. Two different mortar mixtures were made, each with three test tubes which were tested after 7 days. A flexural strength test was performed on three prisms, while a test of compression strength was conducted on all six halves of prisms obtained after the first test, per each mortar mixture. The sample I was made using the ground natural brick sample marked as 1, while sample II was made using the one marked as 2.

According to the Serbian standard SRPS B.C1.018:2015 [167], prescribed for the testing of natural pozzolans, natural calcined pozzolans, and fly ashes, the above-described mixtures of mortars need to have a minimal compression strength of 5.0 MPa after 7 days in order for natural brick to be determined as pozzolanic material. However, various national tests have different value limits. For, example, according to the Canadian standard CSA A3004-E1 for determination of lime reactivity [169], which prescribes the same procedure with small variations, the compression strength of mortar with tested material cannot be less than 5.5 MPa after 7 days, in order to define used material as reactive with lime, and thus pozzolanic [169]. Since all current tests dealing with pozzolanic activity have some shortcomings [170], researchers developed a modified test combining different standards, with the following reactivity classes of potential pozzolanic material according to the compression strength after 7 days: no or very low reactivity (below 2.00 MPa), low reactivity (2.00–5.00 MPa), moderate reactivity (5.00–10.00 MPa), high reactivity (10.00–20.00 MPa), and very high reactivity (greater than 20.00 MPa) [171].

After our testing, mortar mixtures with natural brick I and II showed compression strengths of 3.43 MPa and 5.12 MPa, respectively, while the values of flexural strengths were more closer to each other, that is, 1.37 MPa and 1.77 MPa, respectively (Table 3). Four out of six tested halves of the prisms made with natural brick marked as 2 had values of compression strength over 5.0 MPa, with the highest value at 5.3 MPa; one was slightly lower, 4.9 MPa, and one had a value of 5.0 MPa. Considering the flexural strength of these samples, they were in the range of 1.60 MPa to 2.00 MPa. It is visible that the mortar mixed with the use of sample 1 had lower strengths than the one made with the sample 2, although the sum $SiO_2 + Fe_2O_3 + Al_2O_3$ in this sample was slightly higher.

| Table 3. Mechanica | l features of the mor | rtar samples made | e with natural brick. |
|--------------------|-----------------------|-------------------|-----------------------|
|--------------------|-----------------------|-------------------|-----------------------|

| Sample | Compression Strength (MPa) | Flexural Strength (MPa) |
|-----------|-------------------------------|----------------------------|
| I1 | 3.50 | 1.20 |
| I2 | 3.20 | 1.40 |
| I3 | 3.60 | 1.50 |
| I4 | 3.50 | |
| I5 | 3.40 | |
| I6 | 3.40 | |
| I (avg.) | 3.43 | 1.37 |
| II1 | 5.30 | 1.60 |
| II2 | 5.20 | 2.0 |
| II3 | 5.20 | 1.7 |
| II4 | 5.10 | |
| II5 | 5.00 | |
| II6 | 4.90 | |
| II (avg.) | 5.12 | 1.77 |

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According to the chemical composition requirements in standard ASTM C 618–22 [164], both samples of natural brick had pozzolanic features. According to the gained compression strength using the SRPS B.C1.018:2015 standard, sample 2 (mixture II), had pozzolanic activity (reactivity with lime). It can be determined as moderate according to the abovementioned research. The sample 1 (mixture II) did not fulfil the standard and fell under the category of low reactive materials. In the future, tests can be repeated by monitoring of the mixtures over a longer period, since some materials can exhibit their pozzolanic activity later, and using additional types of analyses [165]. During worldwide research of the pozzolanic activity of naturally fired sediments for the purpose of its use in naturalpozzolana cement for concrete, conclusions were made that they mainly comply with the ASTM standards for natural pozzolans and Portland-pozzolan cement, but that a limited number exhibit good pozzolanic properties in reality and contribute to high early strength. However, those that do not contribute can still be used in those cases where early strength is not a necessity [155]. Research on porcellanite from Trinidad stated that the tensile strength of the mortars prepared with this material using the same ratio of 1:2:9 was much higher than in mortars with volcanic materials from the area of Naples, Santorini earth, and trass. The difference in strength between lime mortars with coarse and fine porcellanites was minimal but still higher in the one with fine pozzolanic material [172] (as cited in [155]).

During the research on pozzolanic features of coal combustion rocks in China created by the metamorphosis of sedimentary rocks, baked rock, sintered rock, and lavalike rock were sampled (from the lowest to the highest firing temperatures) [158]. Total content of silica, alumina, and ferric oxide was the highest in sintered rocks, somewhat lower in lavalike rocks, and the lowest in baked rocks, which confirmed that level of pozzolanicity increased up to a certain temperature, and then decreased, being almost the same in sintered (88.826%) and lavalike (87.234%) rocks and lower (less than 70%) in baked rocks. The pozzolanic activity tests showed that it increased with the smaller particle size, that is, with the greater specific surface area, thus increasing the amount of reactive component. In this context, the highest level was in lavalike rocks, slightly lower in sintered rocks, and relatively low in baked rock [158].

Concrete and asphalt with natural clinker as aggregate were experimentally made in Arizona. The material absorbed too much water and decreased the strength of the concrete, sometimes even weathered out, leaving holes in the concrete, while in the asphalt, it absorbed a large amount of oil [120]. However, in North Macedonia, the material from the bed in the village of Delčevo in the Bregalnica region has been already used in the building industry for the production of concretes [154,173]. During the research of the potential use of natural brick from one of the Kostolac beds (sample 3 in Table 1) [154], experimental mixtures of concrete were made with natural brick as an aggregate, cement CEM I 32.5 as a binder, and water, in a ratio of 1:4.5:1. After 7 days, the average compression strength of the cubes was 6.50 MPa, after 14 days it was 8.625 MPa, and after 28 days it was 12.375 MPa. The values of the static module of elasticity showed that these concretes were much more deformable than classic concrete of the same strength. The thermal conductivity was very good. Considering resistance to frost, the research showed that these concretes should not be subjected to low temperatures and saturated with water at the same time. The natural brick used in this research had many admixtures, sometimes even insufficiently baked clay, or the clay not baked at all, and thus soluble in water (which confirms our assumption that it resembles sample 2). The research suggested the use of the grains above 8 mm for concrete blocks since they are compact and better-baked clay; the remaining grains are proposed for the production of ecological brick, mixing natural brick as an aggregate with pozzolanic features, hydrated lime, a low amount of cement, and accelerators, suggesting these bricks would be successful in the regulation of humidity, while the additives could improve its thermal insulation characteristics [154]. Although conducted on different mixtures, this research can be used in the future studies of the possible function and role of natural brick in different Roman structures.

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Our research was conducted using ground natural brick for the creation of mortar, and thus enabled it to fully react with lime, and other research on concrete used this material as an aggregate, with cement as a binder. However, this research, as well as the research from China, initially confirm that natural brick can potentially be used as a pozzolanic material. Considering lime mortars, the properties of mixtures of lime and pozzolanic material are determined by many factors and their mutual relationships. The pozzolan surface area, its particle size, chemical, and mineral composition, its amorphousness, and the water required for workability all affect its reactivity, and thus mortar strength [174], all of which needs to be taken into account in future laboratory research. One important indicator of the possibility of the use of natural brick as a pozzolanic material in lime mortars can be the practical application of these mortars, which can greatly help in the future conservation of the historical monuments whose mortars, and building materials in general, are scientifically researched.

In the beginning of the 2000s, researchers attempted to utilize natural clinkers in industry, as a coal by product, which is created not only in coal beds, but also in waste dumps with coal refuse. One of those attempts was successful synthesis of zeolites from natural clinkers. The researchers were motivated for this research by the general similarity in chemical composition of natural clinker and volcanic materials (which contain zeolite minerals) [175–177]. The same similarity between volcanic materials and fired ceramics was stated by the researchers of historic mortars [60].

3.5. Experimental Application

Trials of mortar mixtures made with the admixture of crushed and ground natural brick were carried out at the Viminacium Archaeological Park using the traditional method of hot mixing [178]. The process took place during the MoDeCo2000 project international workshop with a conference held in June and July of 2022, named Science for the Conservation of the Danube Limes [14] (Figure 13), led by a building conservator and mason Nigel Copsey [178]. A wall was built using mixtures made with quicklime, Danube sand, zeolite, schist, and natural brick, and their compositions depended on their function in the structure (bedding, pointing, or coping mortar). As building elements, fired bricks, fragments of schist, blocks of natural bricks, and limestone, all once being a part of Roman structures and found in ruins during the excavations of Viminacium, were used, since the aim was to test their contact with newly prepared mortar. During the erection of the wall, three different mixtures were used with the above-mentioned ingredients, according to the function the particular mortar had in the wall (Figure 13).

The structure has been monitored since it was constructed, and its mortars will be tested in the laboratory in spring 2023. The samples of bedding, coping, and pointing mortars will be taken, and their physical and mechanical characteristics will be primarily tested, in addition to their chemical and mineralogical composition. The aim would be obtaining the values of its compression strength and analysing visual features (for the comparison with historical mortars), but also hydraulicity and presence of minerals (for the evaluation of the contribution of zeolite and natural brick as natural materials with pozzolanic features to the composition and properties of mortar). Since, in the mentioned mixtures, the amount of natural brick was not enough to give the reddish colour to the mortar, the results will lead us to further conclusions regarding the possibilities of the use of natural brick in Viminacium mortars by its builders even when we do not see it as abundantly used admixture, as well as to the possibilities of its use in future conservation mortars. The results will be published in the near future, and, along with the other results of the MoDeCo2000 project, they will be a part of the web database, open for the researchers.

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Figure 13. Hot mixing during the MoDeCo2000 workshop. Creating a demonstration mixture (**top left**) and the mixtures for a wall structure using lime mortars, with natural brick as admixture with a photo of the wall 11 days after the building (**top right**, and **bottom**). Photo documentation of the MoDeCo2000 project.

An individual demonstration mixture with greater amount of natural brick was also made during the workshop, using quicklime, natural brick, and sand, in a ratio that broadly corresponds to the one that Vitruvius recommended for lime mortars with river or sea sand, that is lime:crushed/ground brick:river sand ratio of 1:1:2 [71] (II. 5), [179], obtaining the reddish mortar (Figure 13). Although mixing mortars in laboratory conditions is different than what can be expected on site during the mixing performed by masons in the process of conservation, even when the mortar components and their ratio are the same, an attempt was made to initially compare the results of the compression strengths of the mixtures made in the laboratory during the tests on pozzolanic activity and those made during the demonstration on site. After the mixture made on-site was moulded and aged for 35 days in the laboratory, its compression strength was tested, giving a result of 1.85 MPa. The natural

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brick used for the mixture can be comprehended as having similar characteristics as the sample of the natural brick from the bed marked as 2, which was used for the pozzolanic activity test, giving values of mortar compression strength that vary from 4.90 to 5.30 MPa after 7 days.

The differences in compositions of the laboratory and the on-site mixtures were in the type of lime (hydrated vs. quicklime), and, consequently, the method of mortar preparation, the type, purity, mineralogy, and granulation of sand (standard pure quartz vs. fine and sharp building river sand), as well as the granulation of natural brick (ground vs. a mix of coarse and ground). The ratios were also mutually different (1:2:9 vs. 1:1:2), however, the laboratory mixture was prepared using a mass ratio, while during the on-site preparation a volume ratio was used in a way that the masons would use it on site, with no absolute precision in measuring, meaning that an exact comparison of their mechanical features could not be made. Furthermore, in the on-site mixture, about one half of the natural brick was ground and acted as a pozzolanic material, and the other coarse part acted as an aggregate, giving the roughly calculated final ratio binder:reactive pozzolanic material:aggregate for the on-site mixture as 1:0.5:2.5, while in the laboratory mix, all the natural brick (ground brick) reacted with the lime as a pozzolanic material, which helped with gaining strength.

4. Discussion

Natural brick, crvenka, is probably the first raw material suitable for building that Roman soldiers encountered, when they arrived on the Stig plain near the Danube, for the foundation of the Viminacium fortress in the 1st century, apart from wood [46]. It seems that the availability of natural brick made its use very economically advantageous for ancient Viminacium construction activities. However, according to the results of the archaeological excavations, the conclusion can be made that natural brick was abundantly exploited from the bed and used in the 1st century for the building of the legionary fortress, but only as a fragmented material in the second half of the 3rd century and, during the 4th century, for peripheral civilian buildings. Since the recorded use of natural brick in Viminacium buildings dated to the period after the 2nd century is connected with its fragments, it can, maybe, be assumed that those fragments were reused and reworked building blocks of the first Viminacium fortress, which could further support the new assumption that its extensive exploitation could even have stopped after the building of the fortress. The reasons could primarily be sought in its qualities as the main structural building material, which were probably recognised by the Romans as insufficient. However, did they know this from the very start, or only realise it over time, and then started to use other materials more suitable for structural purposes? It is certain that after the 2nd century, we do not encounter blocks made of natural brick in Viminacium buildings, according to archaeological research conducted so far.

This study further suggests that the spatial availability of this material was not that important in the 2nd century and the first half of the 3rd century, which were the most prosperous periods of Viminacium [15], when the schist originated from a quarry in the Ram and Zatonje villages, 15 km away using the water courses, and locally produced fired brick were used. The erection of structures of the excavated building complex of an assumed agricultural character on a site very close to the bed of the natural brick was dated to the period between the 2nd and the 3rd century, a renewal was carried out at the end of the 3rd or the beginning of the 4th century, while at the end of the 4th and the beginning of the 5th century, new buildings were built above a part of the original complex. The structures were built of stone, while a building from the youngest phase was built of fragments of stone, bricks, and tiles [50], as probably cheap and secondary used material. Since natural brick is not mentioned in records, the previous assumption that it was not used for building elements in the prosperous phase of Viminacium because it was not needed might be confirmed, but also that it was not even exploited in the later period, being used in only a very limited number of structures in the suburban zones, as a fragmented and secondarily used material.

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Natural brick has been rarely recorded in the graves or tomb structures of Viminacium [180]. This can possibly be connected to the fact that in the period of its extensive exploitation for the fortress erection, cremation was the prevailing method of burial, and only much later did masonry burial constructions come into the funerary practice of Viminacium. Although the first recorded inhumation burial is dated to the end of the 1st century, only during the 2nd century did inhumation start to develop in Viminacium [180]. Its rise is connected to the second half of the 2nd century, and the final domination happened in the middle of the 3rd century. The building of masonry structures for the inhumated only happened in the very end of this period [181]. Additionally, this period is connected with the most intensive brick production, and thus it was extensively used material for the construction of the graves and tombs.

The research conducted so far has not recorded the use of natural brick in Viminacium city buildings [78,79,182–186]. During his excavations in 1902, the archaeologist Miloje Vasić noted the first phase of the city construction as being made of wattle and daub (from 70 AD to 100 AD to the second phase), the second with the use of schist and brick (the end of the 2nd and the 3rd century), and the third phase with constructions built of fragments of different materials (from the end of the second period to the Hun invasion in the middle of the 5th century) [78,79]. He did not mention any material similar to crvenka, but it cannot be excluded from the different materials in the youngest phase. Additionally, he excavated only a small part of the residential city quarter with workshops. Considering other excavated city public buildings, the use of schist in the urban area started before the end of the 2nd century. The change of the wooden structure of the amphitheatre to a wooden-stone structure happened in the first half of the 2nd century, after which it was incorporated into the city, having previously been a military building [186] (surfaces with crvenka were found during the research of the early phase of the life of the amphitheatre [97,98]). The city baths, where the building phases were dated to the period from the 1st to the 4th century, were built of schist and brick entirely, but its first phase, assumed to be dated to the 1st century, has not yet been investigated [184,185] so as to offer us any data on the used building materials in that period. Since research has been scarcely conducted in the inner-city zone of Viminacium, only future archaeological excavations will give us more information about the building materials used for its construction.

Natural materials with pozzolanic features recorded in historic mortars are those of volcanic origin from the area near present-day Naples, Santorini, or the Eifel region in Germany [60,187], and include tuffs and ashes. The Greeks used the potential of natural and artificial materials to obtain the waterproof quality of plasters and flooring mortars, but its use was not recorded in structural mortars before the Romans [60]. Although both materials can contribute to the formation of strong hydraulic mortars, lime mortars with the addition of natural pozzolanic materials gained ultimate strength quicker, and were thereby preferred for structural purposes [188], as well as for use in marshy and saltwater environments [60], while lime mortars with brick as an artificial pozzolanic material were used in structures in humid and warm environments, as well as in external coatings, due to their high resistance to water penetration [187,188], and were used for flooring, rendering or plastering. On the other hand, the use of both materials in one Roman mortar has been encountered as well [188].

However, in most of the empire, volcanic materials were not available, but the large constructions were still erected, some of which are still standing. The building methods were thus adapted to the locally available materials, and most of the provincial Roman builders created walls with non-hydraulic mortars, although the occasional use of natural hydraulic limes originating from impure limestone, as well as the often negligent use, but also intentional use, of different inclusions, sometimes led to the creation of hydraulic mortars [81]. The type of lime used in Viminacium is currently under research, and the lime remains originating from a Roman lime pit excavated near a Viminacium villa [189], are being investigated. Although is it still not proven that Romans intentionally used hydraulic

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limes, examples were found in ancient Greece, and it was not unusual practice in the Middle Ages [190]. This practice was used in Byzantine as well as Ottoman constructions [191,192].

In the harbour of Caesarea Maritima, builders used pozzolana from the Gulf of Pozzuoli for the exposed structures, but also local artificial pozzolanic material, i.e., combustion residues of organics (ash and charcoal) traditionally used in the area before the Roman conquest, for all other structures, and finally both materials in the most exposed and underwater structures. This confirms intentional use of raw materials for the need of reactive processes in mortars according to the environment [65]. A mix of local and Roman practices, using both ash and charcoal and crushed ceramics, was observed during the research of mortars from the Punic–Roman site of Nora, Sardinia [61].

Terracotta can occasionally be seen in greater amounts in structural mortars in Roman buildings (Hadrian's Wall [193]; hydraulic structures in Le Vieil-Evreux, France [64]; Singidunum fortress, Serbia, researched during the MoDeCo2000 project, etc.), but especially in those used in the early Byzantine period (all samples from the 6th century researched in the MoDeCo2000 project have terracotta in their structure). In these late cases, terracotta dust was used as a highly reactive pozzolanic material, while coarse fragments acted more as a porous aggregate [193,194]. The mortar of Hagia Sophia, with this lightweight aggregate in the wide mortar joints, became a form of concrete according to the researchers [194–196]. However, the research showed that the quality of these mortars was high, due also to the nature of the hydraulic binder which was made of marly limestone or limestone—clay mixtures [196].

Clay minerals and the carbonate content of the raw product have the greatest influence on brick properties after it is fired, but they depend on the firing temperature [197]. The temperatures at which bricks obtain pozzolanic features can be put in the range of between 600 °C and 900 °C or between 450 °C and 800 °C, according to different authors [187,191,198,199], and with the higher temperatures in the mentioned ranges the features are better [107]. It is known that the pozzolanic features of bricks fired at temperatures over 800-900 °C are progressively lower until they become completely nonreactive [191,200]. However, modern research showed that some contemporary bricks made in Britain, Denmark, Lithuania, and Poland, fired at higher temperatures, still retain pozzolanic features up to 1100 °C [201]. According to the experimental research of making bricks using raw materials from modern production in Turkey and Nepal, compression strength gradually increased with increased temperature from 700 °C to 950 °C, after which it significantly decreased, depending on the mineralogical composition. Above a temperature of 1000–1100 °C, the bricks started to melt [202,203]. Most Roman pottery, bricks, and tiles were made with clay that becomes reactive with lime when fired at temperatures of 600–1000 °C, thus, the degree of its reactivity increases from 600 °C to about 930 °C and then starts to decrease [200]. At around 1050 °C, it vitrifies and loses reactivity [200]. Thus, both bricks and pottery can be good pozzolans, except fine ware fired at temperatures at or above 1050 °C [200].

The firing temperature of the researched Roman bricks was estimated to be mostly up to 900 °C (Pergamon [204]), or less, that is 800–850 °C (Romula, Romania [205]). It is, however, known that some bricks could have been fired in Roman kilns at even higher temperatures (Padua, 900–950 °C [206]), depending on their proximity to the fire [207]. Research showed that a brick used as a building element in a Viminacium grave was fired at a temperature higher than 900 °C [107]. In Viminacium, a brick kiln with vitrified, melted, glass-like material, was found [74], indicating that some parts of the kiln reached a much higher temperature [208]. An example of researched pottery kilns in Aventicum (present-day Switzerland) showed that the temperature could have been from less than 500 °C up to 1200 °C, depending on the part of the kiln [207]. The temperature developed in the Viminacium pottery kilns was determined by analyses as at least 850–900 °C and up to 1050 °C [166].

While researching Ottoman mortars from the 14th and 15th century bath buildings in Turkey, the researchers made an assumption that their builders particularly chose brick with

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pozzolanic features for producing hydraulic mortars and plasters [191]. They researched bricks for the construction of the dome, as well as some plasters, and concluded that those used for the construction had poor pozzolanic features, compared to those used for plasters. According to them, this was a consequence of the deliberate use of different materials in the production of bricks made to be used as elements, and those intended for use in plasters [191]. However, we know that the fragments of bricks left after the demolition of a building were reused for the core of later walls, along with debris from stone cutting, broken tiles, and different stones, which were all bonded with lime mortar [179]. There are also records that Romans occasionally used old crushed bricks for the creation of new bricks [209]. Thus, it seems more probable that the Romans recycled many types of bricks and used them also in mortars, but that they could, experientially and based on visual observation, choose those fragments they knew could have better characteristics (pozzolanic), rather than intentionally producing special bricks for mortars. Vitruvius writes about the quality of brick, noting that if it is not made of good clay or it is poorly baked, it is shown immediately when it is exposed to ice or frost [71] (II. 8). Similar doubt in the interpretation of the previous research is shared by Lynne Lancaster, since the material used in mortars could also have originated from tiles or pottery, anything that was cheaper or easier to process, and not only brick [210]. Ancient people indeed had experiential knowledge regarding the quality and characteristics of terracotta considering the firing temperature and raw materials. An the examples of this comes from the pre-Roman period (Iberian), where researchers concluded that the specifically composed pottery was intentionally fired at a specific temperature since the higher temperatures could cause its shrinkage [211].

Considering the mechanical strengths of the bricks depending on the firing temperature [203], possible "overfired" terracotta was probably later rejected by Romans, as having lower values of mechanical properties for use as building elements. We do not know if these elements, fired at a temperature higher than the average limit for obtaining pozzolanic features but less than the average limit for melting (depending on the particular composition), were sometimes used by Romans in Viminacium as mortar additions and admixtures instead, since they could still have pozzolanic features, or if they were rejected and used as a rubble material for the wall cores. Analogously, the choice of natural brick for making building blocks in Viminacium was probably made by the Romans visually, according to its colour, texture, and, thus strength. We encounter dark fragments connected to a natural brick fired at higher temperatures used only for the wall cores, while the blocks were lighter, and probably obtained from material created at lower temperatures.

Some natural brick material could have been created at low temperatures, due to its great distance from the coal fire. Thus, some red fragments in mortars can be both natural and fired brick, no matter of the firing the temperature, except those that are fired at extremely high temperatures, in which case they can be most probably determined as fragments of natural brick. Considering the maximum temperature in a coal bed, the intensity and speed of the fire can be low, when the coal smoulders, but also more than $1200\,^{\circ}\text{C}$ [141]. Combustion-metamorphism generally occurs at high (>600 $^{\circ}\text{C}$) to ultra-high (>1000 $^{\circ}\text{C}$) temperatures, but extreme temperatures can occur in the range from 1500 to $2100\,^{\circ}\text{C}$ close to the coal fire and with fresh oxygen supply [147]. In the Dacian basin of Romania, the temperature range is most often from 250 to 400 $^{\circ}\text{C}$ but can go up to $1200\,^{\circ}\text{C}$ [131,212], while in the Czech Most Basin the temperature of ~980 to $1330\,^{\circ}\text{C}$ was determined [145]. In Australia, the highest temperature of the "Burning Mountain" has been estimated to be $1750\,^{\circ}\text{C}$ [142].

Roman builders, who had already used the terracotta mortars for waterproof lining, were familiar with its higher resistance to cracking during the hardening process, and probably realised that they could also use it in walls [213]. Vitruvius proposed the additional use of one third of ground brick to the mortar mixture of lime and aggregate in the case it is the river or sea sand, and not the pit sand, to obtain "better composition" [71] (V.1). They had empirical knowledge that ground terracotta and natural pozzolana had

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similar properties [213], which can be validated with the citing of Vitruvius [71] (II.6), who connected the features of pozzolana with the effects of fire [213]. The Romans probably accepted terracotta that was obtained by firing as well, as an artificial variety of pozzolana, but it was never in a mass use for structural purposes [213]. According to a researcher, the main reason for this is that mass production of ground terracotta would have had much higher costs than the quarrying of volcanic pozzolanic materials [213]. However, it can be the reason only if this material was obtained as tuff and not ash, since tuff would also have to be further processed in order to obtain fine grains and increase its reactivity for the purpose of use in lime mortars.

Moreover, in those territories with an absence of natural pozzolanic material, builders did not use terracotta massively in structural mortars, thus modifying building methods where needed. Although there are examples that the Romans even added pozzolana to brick mixtures, "it never really replaced ground terracotta" in the mortars that needed to be waterproof, according to Marcello Mogetta, who further adds an interesting assumption that the Romans wanted to preserve the "red hue of the mortar that made it popular in the first place" [213] (p. 32). Conducted laboratory research and experimental work, along with the previously given discussion, can actually speak in favour of the assumption that Romans in Viminacium could have comprehended natural brick as a terracotta-like variety of pozzolanic material of natural origin, actually as a rock whose qualities were changed during the fire effects and used it in mortars. We do not know the appearance of the bed of natural brick near Viminacium at the time Romans started its exploitation. What was the visibility of coal layers at that moment? Were there any fires happening occasionally? Did they know about the nature of the steam if it was coming from the underground? While describing pit sands for mortars originating from different regions of present-day Italy (volcanic, pozzolanic materials, [214–216]), Vitruvius [71] (II. 6) positioned their origin near "springs of hot water" that developed from "the far distant fire and flame" of deep underground heat and the "violent force" of "the Fire Element" [214] (p. 31). There are few cases of lignite combustion in present-day Italy, which created fired clays and porcelanite [133]. In the San Feriolo mine of Ribolla in Tuscany, combustion happened underground at the depth of 60 m [133]. At the site in Colle Fabbri in Umbria, it is still not determined if the porcelanite outcrop is an isolated case created by natural coal combustion, or if it was formed as a result of a small intrusion [217] (as cited in [133])]. This could be similar to the already mentioned misunderstanding of the origin of clinker in USA and its incorrect recognition as volcanic scoria. These are interesting facts for further research as to whether Romans in Viminacium could have understood the nature of natural brick. Did they find it similar to those of volcanic origin, or to terracotta-like materials? It is anyhow the rock whose qualities were changed during the fire effects, as it is the case with both volcanic and terracotta materials.

Thus, ground and crushed terracotta-like materials for Viminacium mortars could have been obtained using real terracotta, but also much easier by using natural brick instead, directly exploiting it from the bed where it is often already crushed and available in large quantities, and thus would cost less than processes connected with fired bricks. It could have been used in later periods also by recycling the already used building blocks. However, except for the flooring, rendering, and plastering mortars, we have encountered only a few structural mortar samples with red admixtures in Viminacium during the project research, moving us away from the assumptions, considering the reasons for the abovementioned justification of use of natural brick as pozzolanic material in mortars connected to the costs compared to the fired bricks.

A part of the Viminacium city with a residential zone was revealed in 1902, and later excavations in the city were conducted in the nearby area, at the sites of the city baths and the amphitheatre. During the excavations of the baths, many scattered mosaic tesserae, small fragments of mosaic floors with tesserae, as well as an in situ remaining part of the floor in one apse, were found [184]. This is the only in situ mosaic found thus far in Viminacium. It encompassed white, grey, and black stone tesserae. Whether its tesserae

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present fired or natural brick, the mosaic fragment observed in this study, as a unique find in Viminacium, is thus very important. Only after future archaeological excavations reveal other in situ mosaic floors or their traces can we make some conclusions on this type of decoration in Viminacium. The connection of the use of shaped natural brick within certain time periods, in case further research reveals that tesserae were made of this material, can be very important in this process.

There can be many possible methods for the use of natural bricks in Roman constructions of Viminacium. Like for other building materials during human history, life of a material included basic process of extraction, processing (production), use (consumption), and disuse (disposal). Although it is not the case in the modern world, this flow was almost always historically widened with reuse, repurpose, and recycling. One of the examples of these processes is use of fired bricks or tiles as building elements, rubble, and mortar admixture in Roman Viminacium. If crvenka as a brick-like natural creation was used instead fired brick during the production of Viminacium mortars, in which we recognize red admixtures, we would be able to comprehend it as a kind of a material that encompassed functions and features of both natural and artificial materials with pozzolanic features.

Laboratory research of natural brick samples, as well as the study in their practical use in mortar, showed promising results on its pozzolanic features. However, only after careful inspection of many Viminacium mortars and their widened research, with different laboratory techniques used, along with the deeper study of natural brick from the bed as well as from Viminacium Roman buildings, can sufficiently justified conclusions on the characterisation of the red mortar fragments as terracotta or natural brick be made.

5. Conclusions

Builders of Roman Viminacium were always focused on the local building materials, commonly using processes of reuse and recycle. The reuse, repurpose, and reworking processes are very visible in its remains, whereby we can encounter fragments of whole building elements originating from old buildings, as well as old building materials in the later structures. The same tradition continued even after Viminacium ceased to exist in the 7th century up to the 19th century. These processes can be observed as a sort of historical sustainability since, before the industrial age, it was usual to use old materials to make something new, "it was the norm in all civilisations," "an evolutionary, additive process," "taken for granted," with "the material resource value to individuals and communities" as "the primary motivation," in a time when "top-down academic interpretations of cultural significance had not been formulated and played no part" [218] (p. 189). The practical reasons for recycling building materials in the Roman period could be numerous. Was there a shortage of any material? Were cheaper and more profitable alternatives sought? Did the value of recycled material change [21]? Was it simply a process considered important for the preservation of nature? "For now, as for the ancient world, recycling and reuse are fundamentally important processes in the economy, and cannot be seen as simply a passive reaction to economic change" [21] (p. 457). Some authors do not believe that careful dismantling of buildings, with individual removal of elements such as bricks, was less intensive than their production, since the old bricks sometimes needed transport from afar [219]. However, by using the manuals from the 19th century, the rates for the demolition of brick walls can be calculated. They show the saving of 51% in man-days for salvage of old bricks comparing to the production of the new ones, while the veneer production was five times harder, and thus very expensive, compared to saving the old panels [220,221].

The natural brick as a local material was present in different life phases of Viminacium, in public and private buildings, having several different functions as a main building material in the early phase, and limited use as fragmented material in later phases. Since its features greatly depend on many conditions connected with its creation, we know that building elements made of it could not be uniform. Did the Romans in Viminacium recognise the features of natural brick and could they predict its behaviour in the building,

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experientially knowing that its wide range of red nuances speaks about the level of its baking, similarly as they did with fired bricks? Its use as shaped building blocks is not recorded after the first period of Roman Viminacium, but its assumed waterproofing and drainage properties were recognised as important for the construction of floors and roads from the very beginning of Viminacium, and even much later in the very landscape, since this practice is used even in modern times.

In this research, a small attempt has been made to get closer to the answer to the question as to whether the Romans in Viminacium used natural brick as a material with pozzolanic features in lime mortars. The possible conclusion that it was used instead of terracotta or along with it, whether as a result of recycling or exploitation, can offer new information for the research of Roman provincial construction, but also for deepening our overall understanding of Roman knowledge about the characteristics and behaviour of different pozzolanic materials in mortars. Conducted laboratory research is only a small part of the scientific procedures that natural sciences can offer to humanities while answering their questions, and can be significantly widened in the future with the aim of gaining more results related to the characteristics of the examined material that could be important for the advance of knowledge in natural sciences connected to materials. In this study, we have tried to present many topics considering construction activities in Viminacium through the archaeological context, using the laboratory research as a companion and a trigger for the development of further research connecting natural and humanistic sciences.

The use of terracotta for making hydraulic mortars throughout construction history can be an example of the recycling process. The same refers to the Roman period, when almost every construction was actually the result of recycling, since fragments of old bricks and stones were also used as aggregate in the Roman concrete [59], as well as in all other wall cores, which is often visible on the very material (brick stamps, traces of use, etc.). If the Romans of Viminacium used natural brick for the same purpose, it would be one more confirmation of the known fact that Roman builders, as well as all other historical builders, created buildings as sustainably as possible. The best question we can ask is whether they ever conducted any construction process as completely unsustainable, with the exception of the examples of luxurious imperial projects and the wishes of wealthy investors. In this case, the proximity of the raw materials and the methods of obtaining them, and thus the cost and practicality of construction with recycling and the reuse of materials, were almost irrelevant. Considering the construction of public buildings where the costs could have been high as well, even if there was no recycling and reuse, it was again sustainable, since use of high-cost or less available materials was conducted only when it was technologically needed. In his writings, Pliny the Elder directly condemns the import of expensive marble for decoration purposes, but also the excessive and expensive exploitation of stone for the same purposes, worrying that "the face of Nature is being everywhere reduced to a level," indirectly appealing for the passing of laws for "forbidding marble to be imported, or the seas to be traversed in search of it"; "Indeed, while making these reflections, one cannot but feel ashamed of the men of ancient times"; "For what utility or for what so-called pleasure do mortals make themselves the agents, or, more truly speaking, the victims of such undertakings, except in order that others may take their repose in the midst of variegated stones?" [222] (XXXVI 1-2).

The conclusions and open questions in this study have shown that this kind of multidisciplinary research can reveal many aspects of the life of Roman Viminacium. We hope that, after future archaeological excavations, accompanied by historical and architectural research, the performance of additional laboratory analyses connected to raw and processed building materials, interpretations based on the cooperation of the humanities and natural sciences, as well as practical work on the use of these materials in building conservation, answers can be found to the numerous questions related to reuse and recycling processes in the construction of Viminacium, and, consequently, about its economy. Sustainability **2023**, 15, 2824 30 of 38

The focus on "how the past was managed in the past" leads archaeology to the insights of anthropological value theories. Through the research on how materials, artifacts, and buildings were reused, renovated, and preserved, but also built over, added to, and, in the end, eventually destroyed, we learn about people's attitudes to the past [223] (p. 84). Therefore, the research of building materials used throughout history in different ways across the Viminacium landscape, where natural brick plays an important role as a specific local material that can be comprehended as one of the carriers of the landscape's immaterial values, can lead us to an understanding of the values people attributed to them, from its first inhabitants until today.

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