Casting metals in limestone moulds

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Abstract

Stone moulds were in common use around the world from the inception of metallurgy until the recent past, and most were certainly used to cast metal directly. Some ancient moulds from the eastern Mediterranean are of limestone and moreover, do seem to have been subjected to heat. However, there has been continuing scepticism amongst metallurgists and metalsmiths that moulds of thermally unstable stone such as these could have received molten copper.

The discovery of a Bronze Age ingot mould from Ras Ibn Hani, Syria, which clearly *had* been used to cast copper oxhide ingots inspired further experimentation into this subject. The mould was described as being of limestone, and casting replications were performed using a special limestone from Gozo, Malta which is used in the construction of ovens. A small oxhide ingot of copper was cast quite successfully, and both mould and operatives survived!

The limestone was shown to be highly fossiliferous and of fairly pure calcium carbonate, very similar to that used for some of the ancient moulds examined from the British Museum's collection and for the Ras Ibn Hani ingot mould, suggesting a long continuity of deliberate choice of this type of limestone through the central and eastern Mediterranean for tasks where heat-resistance was important.

Thus bronze *can* be cast in a limestone mould. However it is suggested that it was only really feasible to cast large simple shapes in single moulds, where loss of surface detail and limited gas generation would not be catastrophic.

Introduction

Stone moulds carved with the negatives of familiar metal artifacts are found all over the world, but were they all used to cast metal directly? The question applies particularly to those moulds carved from a stone which is thermally unstable, of which the most common are the carbonates, limestone and marble. These break down into calcium oxide with the evolution of carbon dioxide at temperatures above 900ºC, and the reaction proceeds quite readily at temperatures in excess of 1,000ºC, which was the usual operating temperature of traditional limekilns (Thorpe and Whiteley 1938, 206). Clearly a mould made of a material which disintegrated with the evolution of gas when molten metal was poured into it would be highly unsuitable, not to say dangerous to use, yet limestone moulds bearing the negatives of shapes such as flat axes, which were normally made of copper or bronze, are frequently attested (Tylecote 1976, 32-3; Tylecote 1987, 211-21). It is noticeable, however, that there is an apparent broad differentiation in the geographic distribution of the use of limestone. Thus there are very few recorded from temperate western Europe, with none at all reported by Tylecote (1986, 82-9) from Britain, apart from the large moulds used in the West Country to cast pewter in the late Roman period (Blagg and Read 1977). In the latter case there is, of course, no problem, because even the highest melting point likely to be encountered (that of lead itself at 327ºC) is still hundreds of degrees below the dissociation temperature of the limestone.

Bronze Age moulds in the Aegean and the eastern Mediterranean

Few limestone moulds have been found in the western Mediterranean; it seems that only in the eastern Mediterranean were moulds of limestone, apparently for casting copper and its alloys, more common, possibly because the limestones found there are more suitable (see below). Thus, although numerous stone moulds dating to the Bronze Age have been found on Sardinia, all are of steatite or chlorite (Becker 1984, Lo Schiavo 1986). Further east the situation changes and, for example, the studies of Branigan (1974, 77-83 and 201-3) for the Aegean and of Catling (1964, 272-5) for Cyprus, both record limestone moulds from Bronze Age contexts. Neither author, however, seems to have appreciated the potential problems in their use, and just assumed that the metal was poured into them directly. Branigan's survey showed that the vast majority of 'open' moulds (that is, single valve moulds) were of stone, and where the stone was identified that the majority were of mica-schist, but limestone was also used. Thus at Troy, for example, Schliemann's (1880, 432-3) excavations produced 15 single valve moulds, 11 of which were of micaceous schist, one was of granite and three were of limestone.

Catling's survey of Bronze Age metalwork on Cyprus produced a rather higher incidence of limestone moulds, with four out of the five stone moulds described as being of soft white limestone. All but one of the latter are now in the Department of Greek and Roman Antiquities in the British Museum. These include the single surviving valve of a two piece mould for casting a socketed spatulate tool, and a single valve mould for billhooks, the same outline being carved on opposing faces of the limestone block. These were both found at Klavdhia on Cyprus (BM GR 1899, 12-29, 91 and 92 respectively) (Fig 1). The third example is of a single valve mould for casting a socketed hammerhead (BM GR 1897, 4-1, 1337) (Fig 2), excavated at Enkomi (Murray 1900). The stone used had previously been part of a much larger mould to cast sickle blades, the remains of two curved indents are preserved on the opposite side to the hammer head. The fourth limestone mould in Catling's catalogue is a fragment of a mould for casting tanged sickle or knife blades, excavated at Enkomi in 1948 and now in the Cyprus Museum (CM 1949 No 678; Dikaios 1969).

The moulds now in the British Museum were examined by us, primarily to confirm the identification as limestone. Examination using a binocular microscope revealed that BM GR 1899, 12-29, 91 and 92 were both of a foraminiferal limestone; BM GR 1897, 4-1, 1337 is a soft fine-grained creamy limestone. The foraminiferal limestone is similar both to the limestone from Gozo, used for ovens etc, which was selected for the experiments described below, and to the Bronze Age ingot mould from Ras Ibn Hani which inspired this research in the first instance.

Both stone and clay moulds were found at the Bronze Age

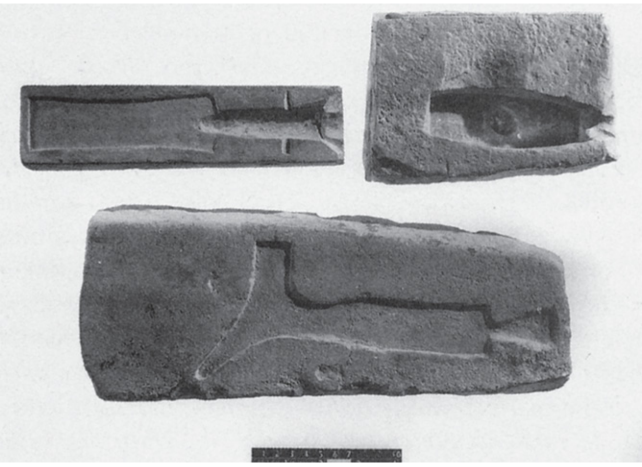

Fig 1: Three limestone moulds from Cyprus now in the British Museum. The triangular shape on the end of the billhook indent could be mistaken for the impression of a putative sprue-filling cup, suggesting the mould was a template for a lost-wax casting. However the indent has no thickness; it would cast as a triangular tab, not a cone.

Fig 2: Limestone mould for casting a socketed hammer. The circular depression in the bottom of the mould would have held in position a clay rod to form the socket space in the casting.

Minoan settlement of Kommos on Crete (Shaw and Shaw 1995, 485-6). The stone moulds are described as being of 'sandy well indurated limestone, similar to that used for the ingot mould at Ras Ibn Hani' (but see the Ibn Hani report below) and were rectangular blocks with bowl-shaped depressions in their upper faces appropriate for the casting of bun ingots. By contrast a series of clay mould fragments (ibid, 506, Reg Nos M 29-40) were found for casting the familiar double-bladed shaft-hole axes by the lost-wax process.

In all of these surveys stone moulds predominated for single valve moulds, with only one two-piece mould recorded as being of limestone. This was for casting the spatulate tool, BM GR 1899, 12-29, 91. In general, however, the limestone moulds were single valve and used to cast large simple shapes without much detail, such as ingots or where it was anticipated that an appreciable amount of hammering and grinding would necessary on the casting to create the finished artifact.

The post-palatial levels at Palaikastro, on Crete, produced only evidence for lost wax casting with clay moulds, particularly for double-bladed shaft hole axes and stands (Hemingway 1996). The led to the suggestion (ibid, 241) that perhaps the stone moulds used during the Palatial period represented mass production to meet the demands of the long distance trade markets, whereas, by contrast, the clay lost wax moulds of relatively simple shapes such as the double axes represented individual production more suited to the restricted local demand of the post-palatial LM III periods. This assumes that the stone moulds were used to cast metal directly, but even if they were used as templates to cast wax positives for investment then the argument still applies because the moulds would still represent mass production, but at one step removed. However the question of the true function of the stone moulds is clearly of importance, and is one which must now be addressed.

Moulds or templates?

Although stone moulds for casting metal were quite common in Europe in the medieval period through to the 19th century, with unequivocal contemporary descriptions of their use in some instances, there has always been some doubt as to the use of such moulds in antiquity; had they really been used directly? Pernice (1904), inspired by the publication of the stone moulds from Troy, carried out a series of experimental replications, casting bronze in a variety of stone moulds. In all cases the experiments were a failure, the castings were imperfect, and the mould was badly damaged whether used hot or cold. On this evidence Pernice concluded that metal had not been cast in stone moulds of any description. On the basis of further work Pernice (1907, 104 and 189, note 21) suggested that the stone moulds were in fact patterns or templates from which wax models had been cast and from which metal castings could be made by the lost-wax method. This is perhaps a salutary example of where the inability of inexperienced modern experimenters to replicate an ancient technology does not necessarily mean that the technology was impossible. Thus, for example, sheets of brass were still being cast in granite moulds in Belgium and the adjacent parts of Germany at least until the end of the 18th century (Galon and Duhamel du Monceau 1764, 102-3).

However, there continued to be considerable doubt amongst those studying ancient metalworking technology as to whether it was safe, or even possible, to cast metal in a limestone mould where gas evolution was a distinct possibility. One of the most feared mishaps in a foundry is for metal to be poured into a damp mould where the resultant evolution of steam either causes the mould to explode, or in some instances has produced a terrifying fountain of molten metal, forced back out of the top of the mould by the freshly generated steam beneath. With visions of this sort of catastrophic gas evolution in mind the use of limestone moulds for casting copper or bronze was doubted, but few, if any, fresh experiments were attempted.

As noted above, Branigan and Catling both assumed the moulds were used directly; Catling going so far as to outline how the casting took place. For the billhooks he postulated that the metal would have been poured into the triangular shape at one end of the mould, which has the superficial appearance of a filler cup, whilst horizontal. The mould would then have been raised to a vertical position against a flat surface to retain the metal, allowing the metal to fill the mould space. Unfortunately for this hypothesis the putative filler cup has no depth and would contain very little metal. It seems more likely that, if this is indeed a mould for direct casting, then the metal would have been poured into the mould and the function of the triangular space, if it was not part of the intended shape of the cast object, was to act as an overflow to stop the metal flooding over the edges of the rather shallow mould. The mould for the socketed hammer from Enkomi can be described as intended for direct casting rather than a template with more certainty. This is because it is difficult to envisage how soft wax could have been released from the deep mould without doing serious damage to it, but a solid metal casting could have been prised out from the pouring channel on the side.

As Pernice had suggested long ago, direct casting is not the only tenable explanation of how the moulds could have been used. The British Museum's label accompanying the limestone mould for billhooks from Klavdhia (BM GR 1899, 4-1, 92), for example, describes it as being a template for casting wax models which could then in turn be cast in bronze by the lost wax method. This explanation was in part the result of the misgivings of the first author of this paper over the feasibility of casting bronze directly, especially in a closed mould. Moreover clay moulds for casting bronze tools by the lost wax method are known from the Minoan period, as exemplified by the clay mould fragments for casting shaft hole axes from Kommos (Shaw and Shaw 1995, 506). However, on removing the Klavdhia mould from its display case in the museum, it became clear that where it had been chipped the stone was red or grey beneath, suggesting that it had been burnt. Furthermore, all three of the moulds now in the British Museum have a fine white surface coating which appears to be powdery calcium carbonate. This may have resulted from the decomposition of the surface of the limestone when very hot. Thus this evidence of strong heating rather suggests that the moulds *had* been used directly after all. There remains the possibility that all three could have been involved in general fires, as unfortunately such notes that survive of their excavation give no indication as to their context. However the fragmentary mould for sickles or knifes from Dikaios' (1969) excavations at Enkomi, mentioned above, is also described by Catling as having a dark grey burnt appearance, and this mould was not found in a burnt layer. Thus the balance of evidence of strong heat on the moulds does suggest that metal was cast in them directly.

The surfaces of the moulds in the British Museum were qualitatively analysed by energy dispersive X-ray fluorescence to determine if any traces of heavy metals were present.The mould surfaces were not cleaned or prepared in any way for these analyses, and there is no record of any cleaning treatments since their excavation over a century ago. No traces of heavy metals could be detected on either the contact surface or the sides of the surviving half of a two piece mould from Klavdia, BM GR 1899, 12-29, 92. On both contact faces of the single valve mould for casting billhooks, BM GR 1899, 12-29, 91, faint traces of copper, lead and tin were detected, but on the sides of the mould only a faint trace of copper was detected. On mould BM GR 1897, 4-1, 1337 from Enkomi a trace of copper was detected on the contact face of the sickle mould, and traces of copper and tin were detected on the break of the hammer mould adjacent to the contact face, which could not be analysed directly because of the geometry of the mould. On the sides of the mould a faint trace of copper was detected. Thus although the amounts of non-ferrous metals detected were in every case small, overall the analyses do provide some support for the argument that metal was cast directly into these moulds.

The discovery of a Late Bronze Age mould for casting oxhide ingots of copper from the site of the Late Bronze Age north palace at Ras Ibn Hani, near Ugarit and Ras Shamra in Syria (Figs 3 and 4) (Lagarce 1986, Lagarce *et al* 1983), was responsible for focusing our attention on this problem in the first instance. The ingot mould, found surrounded by droplets of copper and badly burnt and cracked, had clearly been used for casting oxhide ingots directly. The ingot mould was mentioned in a paper given by Noel Gale and Zofia Stos-Gale at the conference 'Seaborne Trade in Metal Ingots', jointly organised by the Oxford University MARE and the CBA on 16th-17th October 1987. The mould generated great interest, especially as it was claimed to be of limestone. The stone has since been identified as a particularly fine-grained *ramleh*. *Ramleh* is the local name for a shelly limestone found along the coast of the eastern Mediterranean. This particular *ramleh* is harder and finer than the ordinary *ramleh* used for the construction of walls, but the stone is nevertheless porous (Lagarce pers comm, although Shaw and Shaw (1995, 485) describe it as being of a sandy, well indurated limestone). In the discussion which followed at the conference it was suggested, erroneously as it turned out, that perhaps the mould was actually of a siliceous limestone where the majority of the carbonate had been replaced by silicates, thereby rendering the stone much more resistant to heat.

The experiment

One of us, (CD), whilst on the island of Gozo, off Malta, had noted that a certain type of limestone was specially selected for building bakers' ovens and other uses where the stone would be exposed to heat. This heat-resistant limestone was only to be obtained from one stonemason, and apparently it was obtained from just one quarry. Furthermore it was cut from considerable depth to obtain the required grade. To the naked eye this stone was of a closer grained structure, and the fossil inclusions were much less apparent than in the ordinary limestones used locally. In other words it seems to closely resemble the stone of the ancient moulds from Cyprus and from Ras Ibn Hani.

Fig 3: Limestone mould for casting oxhide ingots of copper, excavated at Ras Ibn Hani in Syria. Note the deep channel for running in the molten copper from a furnace (scale 500 mm).

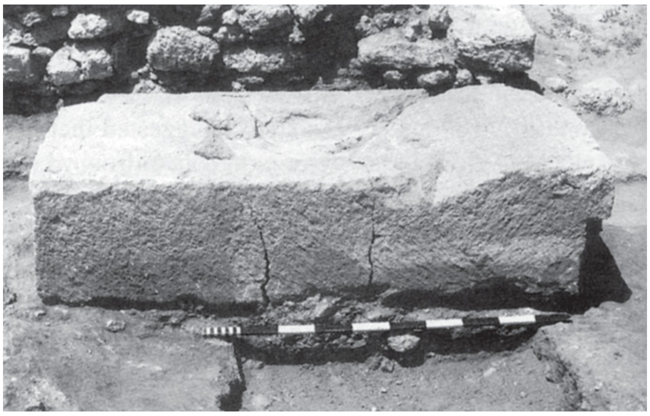

Fig 4: Side view of the ingot mould from Ras Ibn Hani; note the great thickness of the limestone block (scale 1 m).

A piece of this stone was obtained and carved locally to a rectangular block, 360 mm by 275 mm by 95 mm, with the negative of an oxhide ingot at approximately half scale, cut to a depth of 15 mm (Fig 5). The mould was then brought back to England for casting experiments. With no pretreatment other than heating with a blowtorch to dry the contact surface of the mould, an ingot was successfully cast (Fig 6). The thickness of the ingot varies between 12 and 15 mm, with a volume of about 300 ml and a weight of 2.2 kg.

The copper was poured in from a crucible and left to cool, but without covering the top of the exposed molten copper, thereby allowing oxygen from the air to dissolve in the exposed surface of the molten copper. The mould survived the casting, and still remains sound without cracks or signs of use beyond a pink coloration in places where the metal had lain, though the immediate contact surface of the limestone had broken away from the mould to a thickness of about 1 mm and become attached to the undersurface of the ingot, thereby, incidentally, facilitating its removal from the mould. The ingot was extremely porous (as shown by the low density) mainly due to dissolved oxygen coming out of solution as the metal cooled. In the ancient process the solidifying copper would almost certainly have been protected from oxidation by a flat stone lid and/or powdered charcoal. There was evidence of some pinhole porosity on the underside of the ingot, which had been in direct contact with the limestone and this could have been due to carbon dioxide evolved from the limestone.

The surviving ancient oxhide ingots have very characteristic rugged or blistered surfaces, which are sometimes attributed to sulphidic impurities in the copper (Tylecote 1976, 31; Muhly *et al* 1977), or conversely due to the exsolution of dissolved oxygen; Zwicker *et al* (1980, Table 5) noted a correlation between the degree of blistering and the oxide content on the seven ancient oxhide ingots from Sardinia that they had examined. In fact the surfaces do not resemble ordinary gas blistering and they may owe their origin to more mundane causes such as sprinkling water onto the ingot to bring about the rapid solidification of the upper surface, thereby curtailing exposure of the liquid metal to air.

The examination

Samples of two soft, yellowish limestones from Gozo were examined in thin section using a petrographic microscope. The three ancient Cypriot limestone moulds now in the British Museum were not sectioned, but the fracture surfaces were examined under a binocular microscope.

Fig 5: Pouring molten copper into the replica mould.

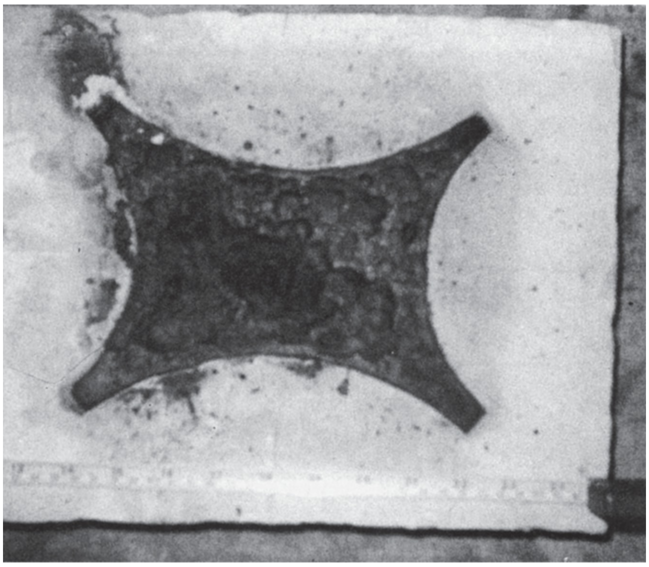

Fig 6: The copper ingot in the mould: note the very porous upper surface (length 213 mm).

The sample from Gozo (BMRL No 29271P), was a limestone used locally for a range of purposes, not specifically related to heating. This limestone consists of abundant foraminiferal microfossils with a microsparrite cement (foraminiferal biosparrite). Sparse grains of fine quartz sand and peloids of glauconite (a potassium iron silicate, micaceous clay mineral) are present. The limestone from the casting mould (BMRL No 29270R) is of a similar type, but has a higher content of fine micritic calcite relative to sparry calcite. It also containes slightly more glauconite, with minor collophane (phosphate). Under the binocular microscope it appears very similar to the limestone used in the ancient Cypriot moulds (BM GR 1899, 12-29, 91 and 92), as noted above. Both of the Gozo limestones and those of the Cypriot moulds are relatively porous (as the ingot mould from Ras Ibn Hani is also reported to be), and this is likely to have contributed to thermal shock resistance and the dispersion of gases evolved by such carbonate decomposition as took place upon heating.

Although the limestone successfully used as a mould in the experiment contains more silicate, in the form of glauconite, than the comparative example, the total silicate contents of both limestones are low. They would not have been sufficient to reduce significantly the effect of heating on the decomposition of the carbonates and the breakdown of the stone.

Attention is drawn to the fact that these limestones, like many others from the Mediterranean region, are relatively recent on a geological timescale. Unlike the hard indurated Palaeozoic limestones of, for example, northern England, they are soft and porous, properties which are likely to have conferred advantages in both working and casting.

Discussion

Clearly it *is* possible to cast copper or bronze in a single valve mould of dry, porous limestone and produce a reasonable ingot. The copper must have been poured at a temperature in excess of the melting point of 1083ºC, although it is likely that this temperature was not much exceeded in antiquity, as the experimentation and examination of the ancient ingots, described below, has revealed. It used to be thought that the metal could have come straight from the smelting furnace without refining (Tylecote 1976, 31), but John Merkel's (1986) experimental work showed that metal from the ingots must have come from a secondary remelting after some preliminary refining, principally to lower the iron content. Casting of metal straight from the smelting furnace was very unusual at any period in the past or present. The *ramo secco* bars from Iron Age Italy are examples of this and are very distinctive, containing macroscopic globules of slag and copper sulphide as well as having very high overall iron contents, quite different from the surviving oxhide ingots (Craddock and Meeks 1987). There was no report of the quantities of smelting slag from the vicinity of the Ras Ibn Hani mould, and this suggests that the copper was smelted elsewhere.

After the metal had been smelted it would have been necessary to refine it by melting in crucibles and stirring, prior to being remelted again and cast. We have no direct information on how this was usually done but the mould from Ras Ibn Hani and some recent experiments provide some clues.

The Ras Ibn Hani mould has a channel cut into the mould and entering at one of the ears (Fig 3). Prof Lagarce suggests that this was to accommodate a crowbar used to lever the ingots out of the mould. This may well be so but the channel seems very deep and flat for that purpose alone, and it seems more likely that its primary function was to channel the metal from a putative furnace. This implies that the metal was run in from a furnace rather than melted in crucibles and poured. If the ingots were regularly made by pouring metal from crucibles then one would expect to find evidence for this on at least some of the ingots in the form of cold shuts where one pour of metal had begun to set before the next could mix with it. In fact no ingots show evidence of interrupted pouring; on the contrary the distinctive upper surfaces suggest that the metal was poured, filled the mould, and set in one operation over a very short period of time.

In his experiments casting oxhide ingots, Merkel used a simple clay shaft furnace, such as we envisage would have been used at Ras Ibn Hani, packed with alternate layers of charcoal and pure copper. This was fired and the copper melted without difficulty. When the melting stage was complete and the liquid copper had collected in the base of the furnace, a hole was knocked in the side and the copper was run through a channel into a sand mould, where it rapidly set.

The ingot was very porous internally, and failed to reproduce the characteristic blistered upper surface found on the ancient ingots. Merkel attributed this to using pure copper rather than the sulphidic 'black copper' of antiquity. The internal porosity was once again due to oxygen dissolved from the air while the unprotected molten surface was exposed to the air.

A metallographic section was cut by Merkel from the ingot which showed large grains, but only a little evidence for the long columnar grains that would be expected if the ingot had cooled slowly, clearly there had not been time for them to develop because the molten copper had not been greatly in excess of its melting temperature when it reached the mould. Merkel also noted that columnar grains were absent on the very few ancient oxhide ingots that had been sectioned for study previously (Balmuth and Tylecote 1976, Muhly et al 1977), and concluded that they too had been poured at temperatures not greatly in excess of their melting temperatures of 1083ºC and thereafter solidified quickly. As the decomposition of limestone only proceeds rapidly at temperatures in excess of 1,000ºC, if this method of casting was used in antiquity there would only have been a very short period, typically of a few seconds duration, during which carbon dioxide would evolve, and then only in the immediate vicinity of the contact surface of the mould, where the temperature would be raised sufficiently for decomposition to take place.

This is confirmed both by the burnt condition of the Ras Ibn Hani mould and by the replication experiment where the immediate surface of the limestone had disintegrated and stuck to the copper.

This does suggest that although it is practicable to cast large simple shapes, such as ingots or flat axes that were then going to be extensively worked by hammering, it would not normally be feasible to cast objects in a limestone mould, where it was intended to reproduce detail from the mould. Similarly, there was almost certainly *some* gas evolution, and although this could be safely accommodated in an open single valve mould, it would be potentially much more dangerous in a closed mould (although note the one Cypriot example described above).

Thus the use of limestone moulds was generally restricted to large simple shapes that could be cast in single valve moulds.

The prevalence of limestone moulds in the Aegean-eastern Mediterranean region was probably due to the presence of large deposits of foraminiferal limestone which provided an easily carved and heat resistant rock that is still used to this day where heat resistance is necessary. The similarity of the limestone used for the moulds from antiquity, as well as the deliberate choice of similar limestones today on both Malta and in the eastern Mediterranean, suggest there has been an unbroken tradition of using this material stretching back at least to the Bronze Age.

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