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Introduction

- 1 Ceramic cooking vessels gain their characteristic properties during manufacture, where the clay-rich paste is transformed by firing to a hard durable ceramic. Usually ceramic vessels can be exposed to elevated temperatures and are strong and tough enough to survive frequent handling. While composition and microstructure of a ceramic depend on its manufacture, they in turn influence a ceramic's material properties and, ultimately, performance. Manufacturing parameters which affect composition and microstructure of a clay-based ceramic include the type of clay, the amount, type, size and shape of aplastic inclusions¹, and firing regime.
- 2 To date, most discussions of archaeological cooking vessels and their material properties have centred on their ability to withstand the thermal and mechanical stresses they are exposed to in every-day use (e.g. Tite & Kilikoglou 2002). Thermal stresses arise for example when placing a cold vessel on a fire, while mechanical stresses can develop through scrubbing or stirring activities. It is only relatively recently that thermal conductivity, a measure for the heat transfer in a material, has been systematically investigated in relation to archaeological cooking ware (Hein *et al.* 2008). The thermal conductivity of a material plays a significant role in the heating efficiency of a ceramic and greatly influences the time required to heat up food (Hein *et al.* submitted).
- 3 When examining the response of archaeological ceramics to mechanical and thermal loads, fracture strength, toughness and thermal shock resistance are considered (Tite *et al.* 2001). Fracture strength determines the amount of force which can be applied to a material until a crack initiates while toughness is a measure of the energy required for crack initiation and propagation. Thermal shock resistance describes a material's ability to withstand sudden changes in temperature and is popular in discussions which

revolve around technological choices in manufacture and the performance of cooking ware (e.g., Tite & Kilikoglou 2002), but also of metallurgical ceramics (Martinón-Torres *et al.* 2006). Unlike strength and toughness, this is not a material property, but a complex parameter, which depends also on the nature of thermal shock (Müller *et al.* in press b). The influence of ceramic manufacture on these parameters in view of archaeological material has been reviewed previously (Tite *et al.* 2001). It has been observed that an increase in firing temperature (Kilikoglou *et al.* 1998) and decrease in amounts of aplastic inclusions (Steponaitis 1984, Kilikoglou *et al.* 1995) results in an increase in *fracture strength*. In contrast, *toughness* has been observed to increase with decreasing firing temperature and increasing amounts of temper (Kilikoglou *et al.* 1995). The influence of temper types and shape on strength and toughness was also studied (Feathers 1989, Feathers & Scott 1989, Hoard *et al.* 1995, Müller *et al.* 2010). Experimental work on the influence of manufacturing choices on *thermal shock resistance* (West 1992: 138) concluded that the 'most effective means of improving thermal shock resistance is the addition of tempering material'. In terms of variability of strength levels upon thermal shocking - high amounts of temper material and low firing temperatures appear indeed beneficial for a ceramic's thermal shock resistance (Müller *et al.* in press b).

- 4 Most studies of utilitarian ceramics which focused on issues related to technology and performance have been preoccupied with investigating the response of the ceramic to mechanical and thermal stresses as outlined above. Discussions of the material properties of archaeological cooking vessels were typically concerned with the suitability of a vessel for cooking as such, assessing, for example, its ability to withstand cycles of heating and cooling and frequent handling. These were assessed either by material testing (e.g., Bronitsky & Hamer 1986, Feathers & Scott 1989, Skibo *et al.* 1989, Hoard *et al.* 1995) or through theoretical considerations (e.g. Rye 1976, Braun 1983). If we consider the requirements that different cooking procedures impose on the material so as to assess the relative suitability of vessels to be used for specific cooking methods, another material property, namely thermal conductivity becomes important and the focus is shifted from absolute considerations towards the affordance of a material for specific cooking processes. Thermal conductivity is linked to heat transfer, a process that is crucial for ceramics employed with heat and which will be discussed in more detail below.

Cooking and heat transfer

- 5 Heat transfer during cooking with the use of a vessel is based mainly on two processes, *heat conduction*, that is the heat transfer in solids or non-moving fluids due to interaction between the molecules or atoms of the material, and *heat convection* which is the heat transfer between a solid or fluid surface and a moving fluid with a different temperature. *Heat radiation*, which is a further heat transfer process, can usually be neglected since temperatures achieved during cooking are still comparably moderate. While different heat transfer processes come into play between vessel and foodstuff, heat transfer within a vessel is determined by heat conduction. Heat conduction is described by the following equation (Kingery *et al.* 1976):

$$\frac{dQ}{dt} = -k \cdot A \cdot \frac{dT}{dx}$$

where dQ/dt is the heat flow through the area A perpendicular to its direction, k is the thermal conductivity and dT/dx the temperature gradient. If the temperatures are not constant, the temperature development is described by the following partial differential equation:

$$\frac{dT}{dt} = \alpha \cdot \frac{d^2T}{dx^2}$$

where α is the thermal diffusivity, which is the ratio of the thermal conductivity k and the heat capacity per unit volume ρc_p , with ρ , the density of the material, and c_p , the heat capacity. Heat transfer within a vessel is thus directed by its material properties, in particular thermal conductivity, but also heat capacity and density, and that is why these material properties are of importance for ceramic vessels exposed to heat.

- 6 A superior thermal conductivity results in an even heat distribution and a pan made of a material with a good diffusivity (and accordingly thermal conductivity) rapidly responds to changes in temperature. In a material with low thermal conductivity, uneven heating creates hot spots, where food can easily be burnt. Materials, in which thermal conductivity and diffusivity are low and heat capacity is relatively high, take longer to reach a steady state condition but they are excellent at retaining the heat once their contents are heated up. It is for their specific thermal properties that today, ceramic vessels are commonly used for slow simmering of stews or casseroles, while materials with higher conductivities are normally chosen to boil or fry foodstuff.
- 7 The second heat transfer process, which has to be considered during cooking, is heat convection. Heat convection emerges between the internal vessel surface and *fluid foodstuff*, such as soup or milk, but also and between ambient air and the outer surface of the vessel as well as between ambient air and vessel contents. Heat transfer by convection can be described with the following equation :

$$\frac{dQ}{dt} = h \cdot A \cdot (T_s - T_\infty)$$

where h is the convective heat transfer or film coefficient of the fluid, A the area of the surface, T_s the temperature of the surface and T_∞ the bulk temperature of the fluid. Apart from the fluid itself, the film coefficient depends on the floating speed, i.e., in the case of the ambient air on the wind speed. In contrast to heat conduction, heat convection is not affected by the properties of the vessel's material. In terms of technical choices, convection can only be controlled through shape parameters, thus affecting the contact area of the interface between the ceramic vessel and the moving fluid. Also important for cooking processes is the heating efficiency η (Hein *et al.* submitted), the ratio between the energy E_{cook} consumed in the actual cooking process and the energy Q_{heat} inserted in the system:

$$\eta = \frac{E_{cook}}{Q_{heat}}$$

- 8 The higher the heating efficiency is, the faster will the foodstuff be heated during the heating phase, and the less fuel is required to maintain temperature once it is reached. Earlier studies have investigated heat transfer through examining what was termed heating or cooking effectiveness (Skibo *et al.* 1989, Schiffer 1990, Pierce 2005). While these studies mainly attempted to assess the influence of surface treatment, i.e. vessel shape and permeability, on heating rates, the thermal conductivity of a material plays a significant role in the heating efficiency of a ceramic and greatly influences the time required to heat up food (Hein *et al.* submitted).

Archaeological cooking ware and cooking methods

- 9 Ceramic clay cooking pots come in many different fabrics: there are differences both in the amount and type of aplastic inclusions present in the clay matrix as well as in firing regimes. So cooking ware from the 3rd millennium BC at Tell Beydar in Syria for example is reported to be relatively low fired with large coarse inclusions (Broeksman *et al.* 2004), as is the cooking ware from the earlier Bronze Age periods at Akrotiri (Müller 2009). Webb and Frankl (2011), on the other hand, note that the Bronze Age cooking ware from Marki in Cyprus, while it is coarser, is also harder-fired than other ceramics, implying that they were exposed to fairly high temperatures. While it appears that in many cases cooking vessels in the Mediterranean during the Bronze Age were coarse and not very high fired, later cooking wares, e.g. from the Roman and Byzantine periods, appear to show a tendency to higher fired, finer pastes. A good example of this is the North African cooking ware that was widely traded in the Roman Empire, particularly during the 3rd and 4th centuries AC, which is high fired and contains relatively fine aplastic inclusions (Leitch 2010, Ikaheimo 2003) As outlined above, these differences in ceramic fabric occur not only over time and space, but also within cooking sets at a site, and reflect differences in raw materials and manufacturing practices employed, such as the addition of particular temper material and the selection of specific firing regimes. Differences in manufacture such as those outlined above may give rise to different material properties of the cooking vessels. This includes their heat transfer properties, which, as noted above, play a crucial role in different cooking practices. While in the archaeological literature the occurrence of new cooking ware *shapes* is frequently linked to different cooking practices, either taken to reflect an influx of new people with distinctive cooking traditions (e.g. Joyner 2007, Ben-Shlomo *et al.* 2008) or changes in dietary practices (e.g. Eerkens 2005), the affordance of the *material* for the different cooking practices postulated has not received as much attention to date.

Case study: Late Cycladic cooking vessels at Akrotiri

- 10 In the following we will illustrate the potential of employing ceramic material properties - and in particular heat transfer - to discuss cooking practices in the Bronze Age site of Akrotiri in Greece. Akrotiri is located in the South Aegean Sea and is an important prehistoric settlement (Doumas 1983). It boasts a remarkable cooking vessel assemblage which is well-represented in successive Bronze Age phases. The Bronze Age cooking ware at Akrotiri shows diachronic variation both in morphology and ceramic fabrics. For the latest phase, Late Cycladic IA (LCIA), two locally produced cooking

vessels have been found, the tripod cooking pot and the funnel-mouthed jar. Examination of the archaeological material which included analyses by thin section petrography, scanning electron microscopy, X-ray diffraction and Fourier-transform infrared spectroscopy revealed that both vessel types were made from the same raw materials, a volcanic clay paste tempered with phyllitic material (Müller *et al.* in press a), but appear to have been fired at different temperatures (fig. 1). The tripod cooking vessels appear to have been exposed to significantly higher firing temperatures than the funnel-mouthed jars, which resulted in differences in microstructure between the two vessel types (Müller 2009, Roumpou *et al.* in press). Since the same raw materials have been used for all locally produced LCIA cooking vessels at Akrotiri, it is the influence of firing temperature on the material's properties on which will focus in the following, before looking at the implications for the particular case study.

1. Thin section (above) and scanning electron micrographs (below) of examples of tripod cooking pots and funnel-mouthed jars.

As can be seen in the thin section micrographs, there are no differences in raw materials used, but tripod cooking vessels appear to have been exposed to significantly higher firing temperatures: the SEM micrograph of an example of a tripod vessel shows a vitrified matrix (left) while a non-vitrified microstructure of a funnel-mouthed jar is depicted to the right.

Influence of firing on material properties

- 11 In order to assess the influence of potters' choices on the material properties of the finished vessels, material tests can be employed. Measurements on experimental briquettes are preferred over tests on archaeological material, due to possible alteration of the archaeological material during use and burial, as well as material requirements. For the present example, the case of Bronze Age cooking vessels at

Akrotiri, we manufactured model materials in order to assess the influence of particular manufacturing parameters on a series of material parameters, i.e. strength, toughness, thermal shock and thermal conductivity (Müller 2009). Results of these have been published, and for a description of materials and test methods employed as well as for a detailed discussion of results, the reader is referred to the relevant publications for strength and toughness (Müller *et al.* 2010), thermal shock resistance (Müller *et al.* in press b) and thermal conductivity (Hein *et al.* 2008).

- 12 As outlined above, when considering heat transfer and cooking methods, it is thermal conductivity which plays an important role, while when discussing the overall suitability of utilitarian vessels, properties such as strength, toughness or thermal shock are frequently employed. Firing to high temperatures results in an increase in strength, while toughness in most cases decreases with increasing vitrification, due to a change in fracture mode and reduced energy absorption during crack propagation in extensively vitrified ceramic matrices (Kilikoglou *et al.* 1995, 1998, Müller *et al.* 2010). Differences, however, are most pronounced in untempered materials, but much less evident in ceramics which contain a relatively high amount of coarse aplastic inclusion, as is the case with many prehistoric cooking vessels. For the same reason, i.e. high amount of aplastic inclusions and subsequent stable fracture mode, it can be argued that also thermal shock resistance was probably sufficient for coarse tempered ceramics when exposed to the temperatures expected for cooking processes (Müller *et al.* 2012, in press b).
- 13 It is well known that the firing strategy greatly influences a ceramic's microstructure. Increased firing temperatures result in increasing vitrification, with calcareous clays developing cellular microstructure, while non-calcareous ones develop dense and less porous microstructure (Tite & Maniatis 1975, Tite *et al.* 1982). Importantly, and as discussed above for strength, toughness and thermal shock resistance, these differences in turn contribute to different physical properties. For thermal conductivity, it has been observed that higher firing temperatures and increasing vitrification of the clay matrix results in an increase of this physical property (Hein *et al.* 2008), an effect which is usually observed for clay-based ceramics (see also García *et al.* 2010). The increase of conductivity is related to the development of vitrification and consequent increasing bonding within the ceramic body; while also increased density and lower porosity contributes. Computer modelling using the finite element methods confirms that the different firing temperatures and microstructures result in materials that would afford the cooking vessels with a different behaviour and energy efficiency (Hein *et al.* in press).

Discussion

- 14 Differences in the manufacture of clay-based ceramics do not only affect strength, toughness and thermal shock resistance, properties which are related to the overall suitability of a vessel to be used in cooking, but also thermal conductivity. High firing temperatures and resultant vitrification of the ceramic body lead to an increase in thermal conductivity. As outlined above, at least in contemporary kitchens, materials with lower thermal conductivity are preferred for simmering food, while for boiling or frying those with higher thermal conductivities are better suited.

- 15 For the case of late Bronze Age cooking ware at Akrotiri, where two different types of locally manufactured cooking vessels were employed for cooking, each fired to a different temperature range, one could therefore contemplate that these two different vessel types - even though they were both used to prepare food - might have been intended or designed for different ways of doing so. In terms of vessel shape, the more open shape of the tripod appears to be better suited to boiling activities, while the more closed funnel-mouthed jar is better for long-term simmering. This is because the heat loss by heat convection is considerably reduced so that the heating energy necessary to maintain the simmer temperature is decreased. One might therefore argue that an adaptation of the cooking vessels in terms not only of their shape, but also of their material properties, namely the adjustment of their thermal properties to specific cooking methods, could have taken place, achieved through firing the ceramic paste to different temperatures. The high firing temperatures which were used in the manufacture of the tripod cooking pot resulted in a higher thermal conductivity beneficial for boiling activities. The funnel mouthed jars on the other side were fired to lower temperatures, which resulted in a lower thermal conductivity better suited for simmering food.

Summary and conclusion

- 16 When examining technological choices in the manufacture of clay-based cooking vessels, it is important to keep in mind that the mechanical and thermal properties of these vessels are connected to their use, the preparation of food. Cooking is inherently related to heat transfer, and the thermal conductivity of a vessel not only influences its overall suitability to be used for cooking food, it also directs the relative suitability of a material to be used for different modes of cooking. For a thorough technical interpretation of the technological variation in ceramic cooking ware production, taking into account the restraints and requirements posed by different cooking practices, a comprehensive examination of ceramic material properties including heat transfer parameters, is a prerequisite.
- 17 The analysis of archaeological cooking vessels in terms of their material properties, ideally coupled with organic residue and archaeobotanical and -zoological analysis, has the potential to support hypotheses such as of the use of different shapes within a cooking ware set for particular types of cooking. Placed in context, the approach presented here is anticipated to contribute to a deeper understanding of food preparation and consumption practices and highlights the importance of integrating different avenues in order to shed light on issues related to culinary practices and diet in the past.
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NOTES

1. Aplastic inclusions, which include for example sand, small rock or mineral fragments, are in many cases naturally present in the sediments employed in ceramic manufacture, but they can also be added by the potter, e.g. to enhance working or drying properties of the plastic clay, or to regulate properties of the final product.

RÉSUMÉS

Cet article examine l'alternance entre les paramètres de la fabrication et les propriétés des matériaux des ustensiles en céramique archéologiques servant à la cuisson. Tandis que les études préalables concernant les vaisseaux en céramique étaient axées sur leur réaction aux contraintes mécaniques et thermiques, l'accent ici est mis sur les propriétés thermiques et leur influence sur l'aptitude d'un vaisseau en vue des différentes méthodes de cuisson. Nous montrons comment les propriétés des matériaux d'un vaisseau sont influencées par sa fabrication et de quelle manière enfin ces propriétés des matériaux peuvent influencer à leur tour la sélection de certains vaisseaux quant aux méthodes spécifiques de cuisson, exemple : les vaisseaux de cuisson d'Akrotiri, Grèce, datant de l'âge du Bronze final.

This paper examines the interplay between manufacturing parameters and material properties in archaeological ceramic cooking ware. While previous studies on material properties have focused on the response of ceramic vessels to mechanical and thermal loads, emphasis is placed here on thermal properties and their influence on the suitability of a vessel to be used for different cooking methods. We illustrate how the material properties of a ceramic cooking vessel are influenced by its manufacture and how these material properties in turn may influence the performance and ultimately selection of certain vessels to be used for specific cooking practices for the example of late Bronze Age (LCIA) cooking vessels from Akrotiri, Greece.

INDEX

Keywords : Cooking ware, archaeological ceramics, thermal properties, thermal conductivity, cooking methods, Bronze Age

Mots-clés : Ustensiles de cuisson, céramiques archéologiques, propriétés thermiques, conductivité thermique, méthodes de cuisson, âge du Bronze

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