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Neutron diffraction analyses of Bronze Age swords from the Alpine region: Benchmarking neutron diffraction against laboratory methods

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ABSTRACT

Bronze Age swords represent the peak of craftsmanship in terms of prehistoric weapon production. Following an extensive study on the manufacture and usage of Bronze Age swords from the Alpine region, we have used non-destructive time-of-flight neutron diffraction to analyse a selection of bronze swords. The diffraction patterns were used to determine the phase compositions and crystallographic textures of the copper tin alloys, in order to reveal information about the working treatment at different parts of the sword blades. Neutron diffraction analyses are benchmarked against results from reference bronze samples, as well as against X-ray fluorescence and metallographic analyses applied to the same objects. Comparing different working intensities of cold working in various areas of the sword blades can lead to information about the usage of the swords as mainly thrusting or as cut-and-thrust used weapons.

1. Introduction

With their complex shape and size Bronze Age metal hilted swords represent the peak of craftsmanship in prehistoric metal casting technology. The manufacture and use of Bronze Age swords has been studied extensively in different regions in Europe, so as for Great Britain (Bridgford, 2000), Austria (Mödlinger, 2011a), Germany (Wüstemann, 2005), Greece and Albania (Craddock, 1976; Koui et al., 2006; Mangou and Ioannou, 1998, 1999), and Slovenia (Trampuž-Orel, 1996). According to the shape of their blades, swords are usually defined as primarily cut-and-thrust weapons, or as primarily slashing weapons. While organic hilted swords seem to be from the beginning a more flexibly-used type of weapon, this is not the case for metal hilted swords. Earlier metal hilted swords show indications for a primarily cut-and-thrust way of use, while newer sword types, in Central Europe usually from Ha A1 (c. 1200 BCE) onwards, were primarily used as slashing weapons. Cut-and-thrust swords are defined by a massive midrib, parallel edges, and a rather short, massive cross-section of the edge, while the more recent, primarily as slashing weapon used swords show broader and significantly thinner cross-sections, with a flat, wide midrib, and a leaf-shaped blade. The blades of metal hilted swords were produced specifically for their main function as cut-and-thrust or slashing weapon, as was demonstrated by Mödlinger (2011b, Mödlinger, 2008). Radiographies of Austrian swords showed that blades of the swords usually interpreted as cut-and-thrust weapons were cast with the haft area on top, so cavities would preferably occur in this area, which is of less importance during the usage of the sword than the tip of the blade. On the opposite, blades for swords primarily used as slashing weapons were preferably cast with the tip upwards, guaranteeing this way a higher casting quality and less cavities in the haft area. The haft area is, due to the junction of hilt and blade by rivets in this very area, already a sensitive part when it comes to using the sword in a slashing mode: the impact energy from the blade when hitting a target would result in significant side stress in the haft area, resulting often in worn- or torn-out rivet holes, or the breakage of the blade. Consequently, casting a blade for a sword primarily used as slashing weapon with the gating not located at the tip of the blade increases the risk of breakage of the blade in the haft area, with less enjoyable consequences for its owner (Mödlinger, 2008).

Nevertheless, we have to bear in mind that every sword may have been used in both stabbing and slashing way, depending on the circumstances during combat. This fact might have been considered also during the production of the blade, i.e. through strengthening the edges by cold deformation. Other indications for the multifunctional use of swords are nicks and cuts on the edges all along the length of the blade, such as use-wear in the central part of the blade for swords primarily used as stabbing weapons, or along the edges in the lower parts of the blade, close to the tip, for swords primarily used as slashing weapons.

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Broken off tips for the latter swords also gives a clear indication of multifunctional use.

To identify post-casting thermomechanical treatment of sword blades, metallography is commonly used. However, to gain information about potentially different thermomechanical treatment in different zones of the blade, metallography is not an option due to its invasive character. In fact, only one such study is known: five samples of the complete cross section of a Middle Bronze Age sword blade from the Thames at Battersea revealed different thermomechanical treatment in different zones of the blade (Coghlan, 1971, 71–73). This is a serious constraint of conventional metallographic analysis: cutting a section from an archaeological object is rarely allowed, and when it is, only one sample can usually be taken.

In combination with one metallographic sample, chemical analysis of the alloy composition, and adequate reference samples, neutron diffraction offers a non-invasive alternative to study the occurrence and intensity of thermomechanical treatment and deformation applied to a metal object in different zones. Time-of-flight Neutron Diffraction (TOF-ND) (Siano et al., 2006; Kockelmann et al., 2006b) can be used to characterise metal in the bulk, without special sample preparation. The bulk composition, texture and microstructure of the metal are determined independent of the corrosion, albeit with limited spatial resolution of a few millimetres. A TOF-ND analysis at a pulsed spallation source has many advantages, such as ease of sample positioning and relatively short data collection times (Kockelmann and Godfrey, in press). Neutron diffraction is sensitive to macroscopic and microscopic phase segregation. For binary alloys with one element in solid solution, element fractions can be obtained by accurate lattice parameter measurements. In the present context, TOF-ND can address one important indicator to interpret swords as either cut-and-thrust or stabbing weapons: the original mechanical working at particular areas along the blade. (NB: others are: the point of balance; the shape of the blade and tang; the points of wear). A non-destructive tool like TOF-ND allows surveying a sword in many different areas of the blade. A multi-point analysis allows a robust interpretation of the sword's technology to be formed.

We analysed ten swords by neutron diffraction on the GEM instrument at the ISIS pulsed neutron source at the Rutherford Appleton Laboratory, Oxfordshire, UK. The diffraction patterns were analysed to determine the structure variations, phase compositions and crystallographic textures on the bronze swords at a multitude of analysis points, to reveal information about the cold-working intensities in various areas of the blades. In this way, information from certain unsampled areas of a blade can be sought that can shed light on the usage of the swords as mainly either thrusting, or cut-and-thrust weapons. Here we present results from neutron diffraction of two swords (Fig. 1), in order to demonstrate the specifics of the diffraction analysis. The results on the entire set of objects will be published elsewhere. Sword 1, which belongs to the Achtkant-type swords, is dated to c. 1400 BCE and was found before 1933 in the river Danube at the Greiner vortex close to St. Nikola, Upper Austria. Sword 2, which belongs to the more recent Riegsee-type (c. 1325–1225 BCE), was found before 1818 in Lorch, Upper Austria. No further details are known about its find circumstances. Both swords are held today in the Natural History Museum in Vienna (Mödlinger, 2011a, cat. Nos. 3 and 15). These sword types – the Achtkant-type and the early Riegsee-type – are considered usually as swords used as stabbing weapons only in classical archaeological literature: the basis for this assumption are typological studies only (see Mödlinger, 2011a, 2011b for further literature). However, known injuries from contemporary Bronze Age skeletons indicate otherwise, as may also specific material treatment, such as hardening of the central edges of the blade, in the case of specific swords. In order to gain more information on edge hardening along the whole length of the sword blade, these two swords were studied not only by classical methods such as metallographic analyses and XRF, but also additional, non-invasive TOF-ND analyses were carried out.

Due to the lack of comparative neutron data on prehistoric bronzes, it is essential to produce and analyse standard samples of known working history that replicate the chemical compositions and possible metal working on the swords. Considering the alloys of the analysed swords, four binary Cu-Sn bronze laboratory sample standards have been measured to compare with the bronze objects, to check reliability and sensitivity of the technique with respect to tin content variations, and for illustration of the neutron diffraction data collection and data analysis steps.

2. Materials and methods

2.1. Metallography

Characterisation of the microstructure and chemical analyses were performed on cross-sections of both sword blades. Samples were taken from the lower third of the blade (sword 1) and from the central region of the blade (sword 2) (Fig. 1). The samples were mounted in cold epoxy resin and polished using various SiC papers (500–1200 grit), and then by diamond suspension paste of up to 0.25 μm granularity. The metallographic structures were characterized by optical microscopy in both bright field (BF) and dark field (DF). The samples were first chemically analysed using XRF, and then etched for metallographic examination using an aqueous ferric chloride solution, Klemm II, and ammonium per-sulphate.

2.2. XRF for alloy composition analysis

A micro X-ray fluorescence spectrometer (μXRF) from Röntgenanalytik Systeme GmbH & Co. KG, model Eagle III XXL, was used for the non-destructive analysis and the determination of the elemental compositions of the bronze blades. The device is equipped with a Rhodium tube and a nitrogen-cooled Oxford Instruments EDAX-system with a Si (Li) detector (FWHM resolution for MnK a = 148 eV). Measurements were carried out on the freshly polished metallographic samples before they were etched. Measurement conditions were: beam 40 kV; μA: 125; Chamber: air; spot size: 0.3 mm; acquisition time/point: 150 s. An average of 20 points per sample was measured (Mödlinger, 2011a, 2011b, 17). The results are listed in Table 1.

2.3. Neutron diffraction for structure and texture analysis

Time-of-flight neutron diffraction (TOF-ND) was performed on the General Materials Diffractometer (GEM) (Kockelmann et al., 2006a) of the ISIS neutron source at the Rutherford Appleton Laboratory, UK. Neutrons are produced by stopping 800 MeV protons in a tungsten target. Fast neutrons enter a liquid methane moderator at 110 K, adjacent to the target and in view from the GEM sample position. The moderator slows down the neutrons which pass through vacuum tubes to the sample position. ISIS is a pulsed source operating at 40 Hz, thus delivering 40 polychromatic pulses of neutrons per seconds to the sample. On GEM the neutron wavelength bandwidth ranges from 0.2 to 3.5 Å, with a maximum of the flux distribution at a neutron wavelength of about 2 Å. The pulsed operation allows determining the wavelength of each diffracted and detected neutron in the polychromatic pulse via a time-of-flight (TOF) measurement. A neutron detector at a scattering angle 2Θ measures the neutron TOF, i.e. the time that passes from the generation of the neutron until its capture in the detector. For a flight path L from source to sample (17 m on GEM) the TOF is related to the interplanar spacings (d): (Kockelmann et al., 2001)

$$d = 1.977 \cdot TOF/(L \cdot \sin\theta) \tag{1}$$

with TOF in milliseconds, L in meters and d in Å. Neutrons scattered by the sample hence produce diffraction patterns, intensity versus d-spacings. GEM has about 7000 individual scintillation detector elements viewing the sample from different angles. To simplify the data analysis