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Testing the accuracy of absolute intensity estimates of the ancient geomagnetic field using copper slag material

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ABSTRACT

The Middle-Eastern copper slag is a promising new material for studying intensity variations in the geomagnetic field with high resolution and precision. The purpose of this study is to test the accuracy of archaeointensity estimates determined using copper slag by addressing two questions: 1) "Does slag material display the magnetic properties required for valid Thellier experiments?" and 2) "What is the accuracy of the archaeointensity estimates derived from Thellier-style experiments on optimal samples?" We address the first question through a comprehensive microscopic and magnetic study of representative archaeological slag samples in order to identify the properties responsible for optimal behavior in Thellier experiments. To address the second question, we reproduced slag samples in the laboratory under controlled magnetic fields and analyzed them using the same IZZI paleointensity technique used for the ancient slag. Microscopic analyses of the archaeological slag show that ferromagnetic phases occur as three-dimensional dendritic structures whose branches consist of submicron-elongated particles. Magnetic analyses show that these dendrites behave as an assemblage of shape-controlled, single-domain-like particles and that their magnetization is thermoremanent. We conclude that slag material can be magnetically suitable for valid Thellier experiments. The laboratory-produced slag material demonstrated similar magnetic and mineralogical properties as the archaeological slag. IZZI experiments showed that non-linear TRM acquisition, even at field strengths similar to Earth's, and TRM anisotropy are important factors to monitor during paleointensity studies of slag material. Anisotropy and non-linearity are probably related to the dendritic shape of the oxide grains. Intensity estimates derived from three laboratory-produced slag samples demonstrated accuracy to within ~5% after applying the required corrections.

1. Introduction

The behavior of the ancient geomagnetic field is one of the few available probes for investigating Earth's deep interior through time. By studying the past strength of the geomagnetic field, or paleointensity, we gain information useful for constraining possible modes of fluid motion in the outer core (Glatzmaier et al., 1999; Selkin and Tauxe, 2000; Johnson et al., 2003; Gubbins et al., 2006; Tarduno et al., 2006). The ultimate aim of paleointensity studies is to establish a well-documented history of geomagnetic intensity changes. Yet, some of the most basic questions remain elusive, such as "How fast can the field change?", and "How strong or weak can the field get?" (Tauxe, 2006; Tauxe and Yamazaki, 2007; Ben-Yosef et al., 2009). Archaeointensity data, covering the past few thousand years, can help answer these fundamental questions by documenting the high-frequency features of the field as recorded by archaeological artifacts. High-resolution archaeointensity investigations are key to documenting short-term events (Gallet et al., 2003; Ben-Yosef et al., 2008a,b; Ben-Yosef et al., 2009; Genevey et al., 2009) and require accurate data that are linked to precise age determinations.

Paleointensity studies frequently rely on the Thellier method (Thellier and Thellier, 1959), which is the most accepted and widely used absolute paleointensity technique (Valet, 2003; Tauxe, 2009). The Thellier method includes a gradual replacement of the sample's original remanent magnetization (NRM) with a laboratory thermoremanent magnetization (TRM) acquired in a known field. This procedure should provide an accurate estimate of the ancient field in which the NRM was acquired, presuming a very strict condition: the sample must retain a TRM carried exclusively by non-interacting single-domain (or quasi-single domain) particles (Tauxe, 2009; Dunlop and Özdemir, 1997).

Accurate paleointensity estimates are very difficult to obtain, mainly because suitable materials for valid Thellier experiments are rare. Most materials contain a dominant non-single domain phase, or carry a significant portion of a remanence of non-thermal origin, such as viscous (VRM) or chemical (CRM) magnetization. In addition, the magnetic properties of the primary remanence carriers may later be chemically or

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physically altered by natural processes or by the multiple heating steps inherent in a Thellier-style experiment. As a result, much effort has been focused on finding suitable materials and demonstrating the validity of the intensity estimates obtained from them.

Ben-Yosef et al. (2008a,b) and Ben-Yosef et al. (2009) recently introduced Middle-Eastern copper slag as a promising new material for archaeointensity studies. Copper slag material is a remnant of the ancient copper industry that flourished in the Middle East for thousands of years starting in the Chalcolithic period (4500 BCE) and extending through the Islamic period (1516 CE). Slag is a waste material from the smelting process, by which copper ores (and fluxes) were heated in a furnace to temperatures of 1200 °C, until a separation between metallic copper and the rest of the melt was achieved. During this process the residual melt lying on top of the copper was removed by letting it solidify in the furnace or by breaking the furnace's walls and letting it pour out. Occasionally, this melt trapped charcoal fragments before solidifying as a slag. In some sites large quantities of slag accumulated in waste-piles. Some of these piles were deposited at a relatively fast rate reaching up to 2 m per century (Levy et al., 2008; Ben-Yosef et al., 2009).

Copper slag has a number of magnetic properties that make it a promising material for archaeointensity studies: 1) The magnetization is thermoremanent with a strong intensity typically on the order of a few A m^2/kg. 2) Rapid cooling in air produces glassy textures with submicrometer grains at the outer margin of the slag, often in the size range of single-domain particles. 3) The cooling rate experienced by the slag after smelting is of the same order of magnitude as the cooling rate applied during Thellier experiments. Therefore, “cooling rate corrections” (Fox and Aitken, 1980; Halgedahl et al., 1980) that compensate for differences in cooling rates are insignificant. 4) Embedded charcoal fragments in the slag enable direct and independent radiocarbon dating of magnetic remanence acquisition. 5) Stratified archaeological cross-sections of slag and charcoals (e.g. Levy et al., 2008; Ben-Yosef et al., 2009) allow for a unique continuous high-resolution intensity record with a potential time resolution of tens of years.

The objective of this work is to thoroughly test the accuracy of archaeointensity estimates determined using copper slag. This study aims to answer two fundamental questions: 1) “Does slag material display the magnetic properties required for valid Thellier experiments?” And 2) “What is the accuracy of the archaeointensity estimates derived from Thellier-style experiments on optimal samples?”. The first question is addressed by a comprehensive magnetic and electron microscopy study of archaeological slag samples that display excellent behavior in Thellier-style experiments. This portion of the study identifies the mineralogy of the ferromagnetic phases, characterizes their magnetic domain state, and finds the magneto-mineralogical qualities of the most successful samples. To test the accuracy of archaeointensity estimates from copper slag we conducted an empirical test using the IZZI protocol on synthetic copper slag samples produced under laboratory controlled sets of magnetic field. The experimental set up allowed us to create samples with similar magnetic properties as in archaeological slag. We analyzed these samples using the very same paleointensity procedures used for the ancient slag in order to quantify the accuracy of the paleointensity procedure in general.

2. Methods

2.1. Archaeological sample set collection

The archaeological sample set for the present study was collected from an exposed section in a 2 m pile of slag, at the excavated site of Timna-30, the Arava valley, southern Israel (see supplementary material, Fig. S1 and Ben-Yosef et al., 2008b for location map). According to excavators, the ages of the exposed sequence span from early 14th century BCE at the bottom, to the 10th century BCE at the top (Rothenberg, 1980). Following Ben-Yosef et al. (2008b, 2009), we classified the slag material in the exposed profile into two groups according to stratigraphic level. The upper part of the pile, labeled in this paper group A, contains massive flat shaped slag, 10 to 40 cm in diameter, and 5 to 10 cm in thickness. Colors in fresh cross-sections are black or dark gray with yellowish and whitish areas. Glassy textures are visible in the rim, and flow textures on the upper side of the slag give it an appearance resembling a small-scaled Pahoehoe lava-flow (see Fig. 7a–f in Ben-Yosef et al., 2008b). The slag fragments in the lower part of the pile, labeled in this paper group B, are irregularly shaped with sizes between 5 and 15 cm. Textures are variable, ranging from glass-droplets to vesicular glass to aphanitic textures (see Fig. 4 in Ben-Yosef et al., 2008b). In some instances, needle-shaped phenocrysts ~ 1 mm long are visible. Slag from both groups show no visible signs of alteration and charcoal fragments are frequently embedded in them.

For a preliminary study, nine samples from the two groups were collected from different relative heights (Fig. S1 in supplementary material) and were analyzed using an absolute paleointensity IZZI (Tauxe and Staudigel, 2004) protocol. The five samples showing the best behavior in the IZZI experiments were selected for further analyses and are reported in this study. The sample set consists of one sample from group A (IS26C) and four samples from group B (IS26E-G, I). A further paleointensity study of site Timna-30 including an archaeological excavation designed especially for high-resolution sampling of slag is currently in progress.

2.2. Re-melting experiments

The purpose of the re-melting experiments was to empirically quantify the accuracy of paleointensity estimates derived from the slag material using the IZZI protocol. The experiments were carried out in the experimental petrology laboratory at the Institute of Earth Sciences, Hebrew University of Jerusalem. The experimental setup built for this study is shown in Fig. S2 in the supplementary material. Starting material was a crushed slag from site Timna-28, located a few kilometers south of site Timna-30. The crushed slag was put in laboratory-built crucibles made from a fire-resistant brick material machined into cylindrical shapes with an inner diameter of 1.5 cm and a height of 2.5 cm. A thermocouple was assembled on the bottom of the crucible so that sample temperatures could be digitally recorded throughout the experiments. The crucible was fixed on an alumina rod and inserted into a Carbolite 1-atm tube furnace. The rod allowed us to lower the crucible from the furnace's hot spot to a cooling chamber located below the furnace. The cooling chamber was built as an extension of the furnace's tube. A system of three-axis Helmholtz coils, connected to three DC current sources, was used to control the magnetic field inside the cooling chamber. The field in the cooling chamber was measured in four positions using an Applied Physics APS-520 three-axis fluxgate magnetometer, thereby allowing estimation of uncertainty in the field.

Preliminary experiments showed that the crushed slag melts completely after 1 h at 1300 °C. Quenching the melt by rapid cooling outside the furnace yielded a glassy material with very weak and unstable magnetic signal. Scanned Electron Microscopy (SEM) analysis of the glass showed no visible crystals. Additional experiments showed that different types of crystals form when slower cooling rates are applied between 1300 °C and 1000 °C. The final and most suitable experimental procedure found in this preliminary work included three stages (supplementary material, Fig. S2): First, a crushed archaeological slag was heated to 1300 °C for 1 h in order to guarantee complete melting. Then, the melt was cooled to 1000 °C using a controlled, constant cooling rate. Finally, when the temperature reached 1000 °C, the sample was lowered to a cooling chamber below the furnace, into a predetermined stable magnetic field.