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Studies on rock magnetic and paleointensity of some archaeological artifacts from Tamilnadu, India

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Abstract: This study investigates the magnetic mineralogy and paleointensity values of a collection of archaeological artifacts (pottery). The actual magnetic carriers and their domain states present in the archaeological pottery were obtained using the low field susceptibility, thermomagnetic curves and acquisition of isothermal remanence. The magnetic mineralogy of all the samples was dominated by ferrimagnetic mineral (magnetite/magnetite with low titanium content), which was suitable for paleointensity measurements. The geomagnetic paleointensity value obtained by subjecting them to modified Thellier and Thellier method, is found to be (48.81±0.15) μT.

Key words: Magnetic susceptibility, Archaeological pottery, Paleointensity
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INTRODUCTION

Archaeomagnetic studies have undergone extensive development during the last few decades to reveal information about the long-term behavior of the Earth's geomagnetic field and, when an adequate reference curve exists, can date archaeological artifacts. The necessary condition for the suitability of archaeomagnetic investigation is that the archaeological artifacts must be heated to high temperature, i.e., Curie temperature (T_c) of the respective minerals, which fossilizes the Earth's magnetic field and its direction at the time of last firing. The measurements of remanent magnetization allow the determination of the direction and intensity of the Earth's magnetic field at the moment of cooling the burnt structure. The geomagnetic field can be obtained from those remains found "in situ" since last firing (clay plasters, burnt soil and in some cases of bricks). The other baked-clay finds (pottery, bricks) can give information about the paleointensity, but not the direction. The type of magnetic minerals (remanence carriers), grain size and domain states are also important factors in determining the reliability of the results possessed by the artifacts. Magnetic studies on archaeological artifacts are usually focused on obtaining the ancient direction and intensity of the geomagnetic field at the time of last burning (Abrahamsen, 1996; Kovacheva *et al*., 1998; Jordanova *et al*., 2001; 2004; Herries *et al*., 2007; Casas *et al*., 2007; Gómez-Paccard and Beamud, 2008;). In the present study, the rock magnetic properties and paleointensity measurements are made on recently excavated pottery from Thandikudi (TDI) (12°35′ N, 76°53′ E), Dindigul District, Tamilnadu, India.

METHODS

Mass-specific magnetic susceptibility at low frequency (χ_{LF}) and high frequency (χ_{HF}) was measured for cylindrical shaped samples with the

Bartington MS2B dual frequency meter at two frequencies (χ_{LF} at 0.47 kHz and χ_{HF} at 4.7 kHz) with measuring accuracy of 1×10^{-5} SI unit by applying the field strength of 80 A/m. Percentage frequency-dependent magnetic susceptibility $\chi'_{FD}=(\chi_{LF}-\chi_{HF})/\chi_{LF}$ and mass specific frequency dependent susceptibility $\chi_{FD}=\chi_{LF}-\chi_{HF}$ were then calculated. The difference between the measured magnetic susceptibility at low and high frequencies depends on the concentration of the grain having relaxation frequencies in this interval. The parameters χ_{FD} and χ'_{FD} are used to detect ultrafine (<0.03 μm) ferrimagnetic minerals lying in the superparamagnetic (SP) grain size. High-temperature behavior of magnetic susceptibility was studied using MS2 temperature-susceptibility system (Bartington Instruments Ltd., UK) equipped with a high-temperature furnace MS2WFB. The temperature dependent susceptibility measurements were performed by keeping the powdered samples in a quartz tube and heating it up to 700 °C and cooled down to 100 °C, by recording every 2 °C interval. The accuracy of a single susceptibility measurement is 1×10^{-5} SI unit. Isothermal remanence acquisition was carried out using impulse magnetizer (pulse magnetizer IM 10~30, ASC Scientific, USA) and the magnetizations were measured with MINISPIN spinner magnetometer (MOLSPIN Ltd., UK) with an accuracy of 2.4×10^{-6} A/m. The samples were subjected to the modified Thellier and Thellier (1959) method to obtain the paleointensity values. Thermal demagnetization was carried out using magnetic measurements thermal demagnetizer (MMTD, UK) followed by intensity measurements using MINISPIN spinner magnetometer.

RESULTS AND DISCUSSION

Susceptibility

Magnetic susceptibility measures the 'magnetizability' of a material in the natural environment, which mainly tells us about Fe-bearing minerals that are found in soils, bricks, rocks, dusts and sediments (Thomson and Oldfield, 1986). Magnetic susceptibility (χ) describes the magnetic response of a sample when exposed to a (generally weak) magnetic field. χ is mainly a function of the concentration and mineralogy of the ferrimagnetic (magnetite, maghemite, Fe-sulphides) minerals present, but can also depend on the strength of the applied magnetic field and the particle size distribution of the magnetic grains. In the absence of ferrimagnetic minerals χ can be due to antiferromagnetic, paramagnetic and diamagnetic minerals. Magnetic susceptibility χ is also dependent on sample size. Therefore, it is customary to present susceptibility as mass normalized susceptibility χ (Mooney *et al*., 2002). The magnetic susceptibility value suggests the presence of ferrimagnetic mineral concentration (Thomson and Oldfield, 1986; Dunlop and Özdemir, 1997).

Magnetic susceptibility and its frequency dependence are widely used in studying magnetic enhancement and are also useful in the detection of fine magnetite/maghemite grains (Mullins and Tite, 1973; Maher, 1988; Dearing *et al*., 1996a; 1996b). The frequency dependent susceptibility is a non-destructive indicator to decipher the nature of magnetic carriers that is very much suitable for archaeomagnetic studies. Dearing *et al*.(1996a; 1997) have reported that burnt clay samples with $\chi'_{FD}>2$ have detectable concentration of SP grains, and if χ'_{FD} is around 6~10, samples contain significant amount of fine SP grains of size 0.012~0.023 μm. Hunt *et al*.(1995) reported that a sample containing significant fraction of SP grains (near 20 nm in magnetite) will thus have a high value (up to about 12) of χ_{FD}. Dearing *et al*.(1997) showed in a model mixing experiment that addition of increasing amount of multidomain -magnetite grains to soil, containing predominantly SP magnetite grains ($\chi'_{FD}=10.5$), causes χ'_{FD} to decrease to <2, while χ_{LF} increases with concentration. Table 1 gives the range of magnetic parameter in the present study. Magnetic susceptibility χ_{LF} of pottery samples are more evenly spread over an interval 9×10^{-7}~61×10^{-7} m^3/kg pointing to higher magnetic enhancement. The high χ_{LF} values of the samples are due to higher firing temperature achieved during baking. High values of χ_{FD} indicates the presence of very fine grained metastable magnetic grains spanning the SP-stable single domain (SSD) boundary (Eyre, 1997; Worm, 1998). All the samples show $\chi'_{FD}>2$ and most of the samples fall in between 4~9 indicating the significant content of SP magnetite grains. Fig.1 shows the χ_{LF} and χ'_{FD} for the samples numbered from TDI-1 to TDI-15. The results suggest that the pottery samples under investigations are magnetically enhanced materials in terms of concentration and degree of crystallinity of ferrimagnetic mineral magnetite.

Table 1 Mineral magnetic parameters of archaeological artifacts (n=15)

Magnetic parameter	Range
χ_{LF} ($\times10^{-7}$ m^3/kg)	8.630~60.530
χ_{FD}	2.010~4.020
χ'_{FD}	4.960~8.121
H_{cr} (mT)	30~40
S-ratio	0.722~0.990
Q-ratio	1.032~5.513
T_c (°C)	570~590

H_{cr}: remanence coercivity; S-ratio: $S_{100}=IRM_{-100}/SIRM$; Q-ratio= NRM/$\chi_{LF}\times0.5$ Oe; T_c: Curie temperature

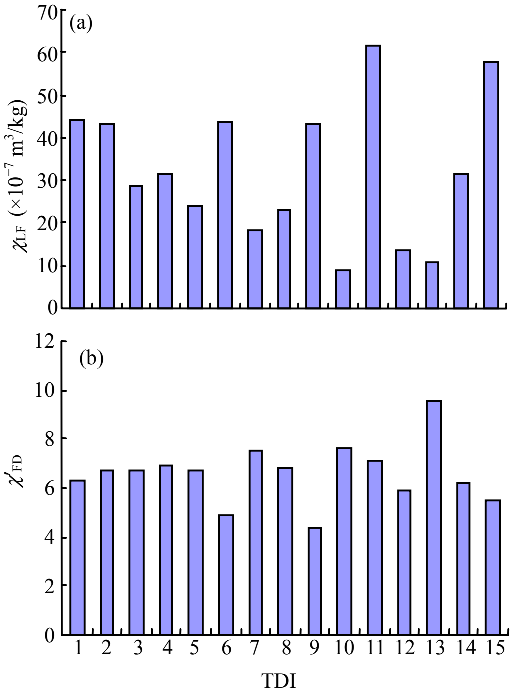

Fig.1 (a) χ_{LF} vs archaeological artifacts and (b) χ'_{FD} vs archeological artifacts

The temperature-susceptibility measurements is to monitor the variations of the susceptibility in order to determine the Curie temperature and to identify magnetic minerals which are responsible for acquiring ancient geomagnetic field and nature of mineral transformation, if any. The heating and cooling curves also reveal chemical and structural changes that can occur as a result of thermal treatment (Atkinson and King, 2005). High-temperature behavior of magnetic susceptibility has been studied in order to determine the main magnetic minerals, responsible for magnetic enhancement. Jordanova *et al.*(2001) reported that the enormous increase in susceptibility values during cooling is most probably due to breakdown of clay minerals and formation of new strong ferrimagnetic phase, indicating that these materials are not burnt to high-temperatures during baking. Samples with basic reversibility of heating and cooling curves suggest no mineralogical change during heating, which are durable for paleointensity studies. The thermomagnetic analysis of magnetic susceptibility is a powerful method for detection of hematite, even when it is present in minor quantity. The main reason is that after heating the material above 580 °C, magnetite does not contribute any more to the measured susceptibility signal therefore allowing detection of hematite. Thermomagnetic curves for representative pottery samples are shown in Fig.2. All the samples show the Curie temperature T_c to be around 580 °C, characteristic of ferrimagnetic mineral magnetite/magnetite with low titanium content. The reversibility of the heating and cooling curves suggests that the pottery samples might have been burnt well above the Curie temperature and there is no further change in mineral phase.

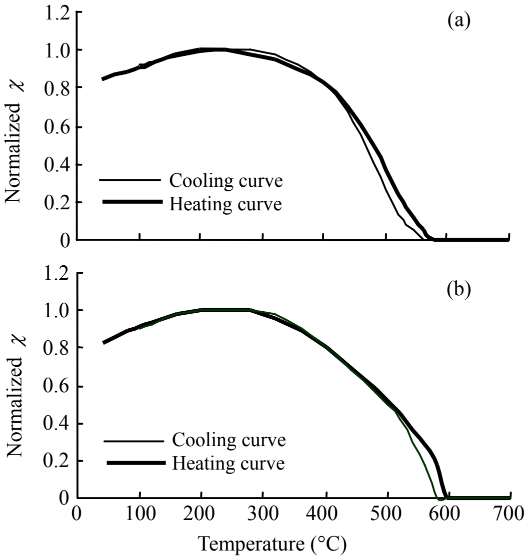

Fig.2 Temperature dependent magnetic susceptibility of (a) TDI-9 and (b) TDI-12

Isothermal remanence

Isothermal remanent magnetization (IRM) is acquired by a sample after exposure to, and removal from a steady direct current (DC) magnetic field. IRM depends on the strength of the applied field, function of magnetic mineralogy and also grain size. The maximum remanence that can be produced in a sample is saturated isothermal remanent magnetization (SIRM). IRM is often used as an indicator for the ferrimagnetic minerals, and antiferromagnetic minerals such as haematite and goethite are also capable of acquiring IRM (Mooney *et al*., 2002). After a sample has acquired an IRM, it is often possible to (partially) demagnetize the sample by exposing it to a magnetic field in reverse direction. Such a partial demagnetization can yield information about the ease of remanence acquisition or the coercivity of the sample. Alva-Valdivia *et al.*(2003a) reported that in IRM acquisition curves the saturation reached at low/moderate fields of 150~200 mT indicate the ferrimagnetic phase corresponding to some (titano) magnetites or (titano) maghemites. If the saturation is not reached even in the maximum available field, the behavior belongs to high coercivity mineral (titano) hematite and (titano) maghemite. In the present investigation the occurrence of saturation below 300 mT and remanence coercivity (H_{cr}) at about 30~40 mT for all the samples indicate the magnetite, probably to be the main magnetic remanence carrier (Zhu *et al*., 2000; Tian *et al*., 2002; Lu *et al*., 2005). The *S*-ratio ($S_{100}=IRM_{-100}/SIRM$) where IRM_{-100} denotes an IRM acquired in a reverse field of 100 mT after acquiring SIRM, can be used to gain information about the magnetic mineralogy (Bloemendal *et al*., 1992). *S*-ratios close to +1.0 are indicative of ferrimagnetic minerals, while low *S*-ratios (<0.6 or even <0) are caused by the presence of antiferromagnetic minerals. IRM acquisition curves give information about the coercivity distribution of a sample, indicating the field at which a sample acquires its remanence. In the present study, all the samples show *S*-ratio values>0.6 which reflects the presence of ferrimagnetic minerals.

It is also worthwhile to examine the magnetic parameter Koenigsberger's-ratio (*Q*-ratio=NRM/χ_{LF} ×0.5 Oe) which gives the type of mineral and its domain state that produce a dominantly induced remanent magnetization and the value 0.5 Oe corresponds to a magnetizing force of 39.79 A/m (McEnore *et al*., 2001). The high *Q*-ratio values are characteristic of stable (thermoremanent) origin of natural remanent magnetization (NRM) while low values (*Q*-ratio<1.0) are for other non-stable remanence (Dunlop and Özdemir, 1997). The *Q*-ratios provide a relative importance of remanent and induced magnetization, with the remanence being dominant for *Q*-ratio>1.0 (Alva-Valdivia *et al*., 2003b). Variations in remanent intensity and susceptibility depend on volume content of magnetite. The *Q*-ratios>1.0 indicate the presence of single domain or pseudo single domain magnetite grains in all the samples and suggest that samples are suitable for archaeomagnetic analysis. The IRM acquisition curves for some representative samples (e.g., TDI-9 and TDI-12) are shown in Fig.3.

Paleointensity measurements

The Earth's magnetic field varies in direction and intensity over historical time scales, a feature that is thought to be generated by the non-dipole field components of the geodynamo. The Palaeointensity fossilized in the baked materials can be retrieved through archaeomagnetic investigation. Most paleointensity determinations using archaeological artifacts and lava flows are based on the classical Thellier and Thellier (1959) method or its modification (Coe, 1976; Kono and Ueno, 1977). The modified Thellier and Thellier method involves heating the sample in a zero field to a number of increasing temperature stages (100 °C to 600 °C in steps of

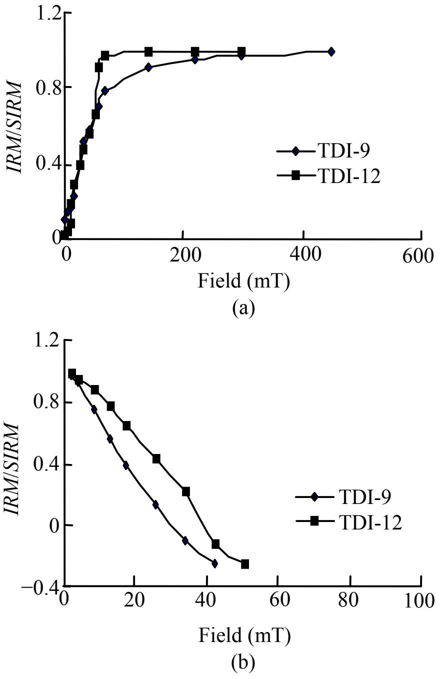

Fig.3 (a) IRM acquisition and (b) Back-field DC demagnetization curves

100 °C) and measuring the intensity of magnetization remaining after each stage is NRM. Once all the magnetization is removed, the process is repeated but with the sample exposed to a known reference magnetic field during heating and the measured magnetization is thermal remanent magnetization (TRM). For the measured magnetization values (NRM and TRM) the Arai plot has been drawn. The thermal demagnetization curve and the remagnetization curve together give the so-called Arai plot, as shown in Fig.4. The slope of the best fitting line to the linear part of the Arai diagram multiplied by the value of reference field (laboratory field) gives the intensity of ancient geomagnetic field $B_{\mathrm{anc}}=(NRM/TRM)\times B_{\mathrm{lab}}$ (μT), which is the value of the field at the time of last burning.

The paleointensity value of all the samples is found to be (48.81±0.15) μT. This value of B_{anc} was fixed in the secular variation curve drawn by Ramaswamy and Duraiswamy (1990) and found to be in the period 300 before Christ, coinciding well with the dating suggested by archaeologist.

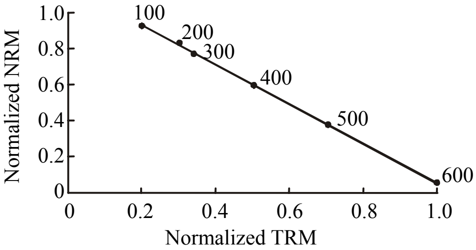

Fig.4 Arai diagram of Thandikudi pottery TDI-9

CONCLUSION

(1) The higher values of mass specific susceptibility obtained mainly depends on the Fe-sources available from the parent unbaked clay as well as on the higher firing temperature achieved during baking. The higher firing temperature during baking is also revealed from the high temperature behavior of susceptibility with Curie temperature of around 580 °C, probably magnetite as the main ferrimagnetic mineral. The reversibility of the heating and cooling curve reveals the suitability of the samples for paleointensity measurements.

(2) The χ'_{FD} values falling in between 4~9 indicates that the samples contain significant amount of SP magnetite grains. IRM acquisition curves with saturation at fields below 300 mT and coercivity value 30~40 mT indicate the ferrimagnetic phase. The *S*-ratio>0.6 is indicative of ferrimagnetic mineral and *Q*-ratio>1.0 reflects the presence of single domain or pseudo single domain magnetite grain in all the samples.

(3) From the above rock magnetic study it is evident that all the pottery samples have stable remanence and the B_{anc} value is found to be (48.81±0.15) μT by using modified Thellier and Thellier (1959) method. The inferred age obtained from this study is consistent with the archaeological evidence available.

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