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THE USE OF MINERAL MAGNETIC PARAMETERS TO CHARACTERIZE ARCHAEOLOGICAL ARTIFACTS

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This study investigates the magnetic mineralogy of a collection of archaeological potteries. Actual magnetic carriers and the domain states of the constituent magnetic fine particles have been obtained from the acquisition of isothermal remanence and low field susceptibility measurements. The magnetic mineralogy of all samples has been dominated by ferrimagnetic mineral (magnetite/magnetite with low titanium content) which is suitable for paleointensity measurement in determining the intensity of the ancient geomagnetic field.

Keywords: rock and mineral magnetism, archaeological pottery

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1. Introduction

Archaeology deals with the systematic study of the relics of the ancient past and throws light on the life and cultural development of a race. The relics are remains, in general, of buildings, burial places, implements, utensils, and ornaments belonging to periods about which there are no written records. Excavation at an archaeological site may reveal the depth of civilization. The unearthed clayware, broken statues, ceramic sherds, corroded armour, weapons, etc., give valuable information about the material environment during the ancient times.

Archaeomagnetic studies have undergone an extensive development during the last few decades to reveal information about the long-term behaviour of the Earth's geomagnetic field, and when an adequate reference curve exists, it can date archaeological artifacts. The necessary condition for the suitability of archeomagnetic 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).

Detailed mineral magnetic investigations have been carried out on archaeological potteries in order to identify the minerals present, which are responsible for the record of ancient geomagnetic field. As no single method can provide complete and accurate information on the mineral magnetic properties of the sample, several techniques have been used in this study. The type of magnetic minerals (remanence carriers), their concentration, and domain states are important factors in determining the reliability of the results found in the artifacts [1–4]. The artifacts are subjected to rock magnetic studies like magnetic susceptibility (frequency) and isothermal remanent magnetisation (IRM) acquisition in order to characterise the main magnetic phases and to select the most suitable samples for paleointensity measurements.

2. Site and methods

Bhon (BON), lat. 76° 39′ E, long. 20° 55′ N, is situated at Sargrampour Taluk of Buldana District, Maharashtra, India and the excavations have been carried out by Deccan College, Pune, India. Representative

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samples from different trenches at various depth were used for the present study.

Mass-specific magnetic susceptibility at low frequency (χ_{LF}) and high frequency (χ_{HF}) have been 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 a measuring accuracy of $1\cdot10^{-5}$ SI unit by applying the field strength of 80 A/m. Percentage frequency-dependent magnetic susceptibility $\chi_{\mathrm{FD}}\% = (\chi_{\mathrm{LF}} - \chi_{\mathrm{HF}})\cdot 100/\chi_{\mathrm{LF}}$ and mass specific frequency dependent susceptibility $\chi_{\mathrm{FD}} = \chi_{\mathrm{LF}} - \chi_{\mathrm{HF}}$ are then calculated. The difference between the measured magnetic susceptibility at low and high frequencies depends on the concentration of the grains having relaxation frequencies in this interval. The parameters χ_{FD} and $\chi_{\mathrm{FD}}\%$ are used to detect ultrafine (<0.03 μm) ferrimagnetic minerals lying in the superparamagnetic (SP) grain size. Anhysteretic remanence magnetisation (ARM) has been produced by a direct field (50 μT) and a maximum alternating field (AF) of 100 mT. AF demagnetisation has been performed on a laboratory built tumbling AF demagnetiser and IRM using a pulse magnetiser model MMPM9. Remanences are then measured with an Agico molspin spinner magnetometer.

3. Mineral magnetic study results and discussion

3.1. Frequency dependent 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 [5]. Susceptibility itself depends upon the concentrations of ferrimagnetic grains (mainly magnetite). Magnetic susceptibility χ is also dependent on the sample size. Therefore, it is customary to present susceptibility as mass normalized susceptibility χ [6]. Measurements of frequency-dependent magnetic susceptibility (difference between magnetic susceptibility measured at low and high frequency of the inductive magnetic field) are now widely used for detection of fine magnetite/maghemite grains in soils and rocks [7–11]. The basis of this technique is the Neel's theory for superparamagnetic relaxation [12] of fine particles, which have relaxation times lying between the two measuring frequencies. The difference between the measured magnetic susceptibility at low and high frequency depends on the concentration of the grains having relaxation frequencies in this interval. Mass-specific and percentage frequency dependent susceptibility are the two parameters most frequently used.

Generally, several factors play the most important role in determining magnetic enhancement of fired clay. These are the properties of the initial unburnt material through its specific iron content (Fe incorporated in clay minerals or Fe-oxide/hydroxides); the degree of burning which in most cases depends on the kind of the remains (e. g. fired clay, brick, oven) and determines the final magnetic mineralogy of burnt clay materials [1]. In the present study pottery samples from Bhon have been subjected to range of rock magnetic measurements in order to elucidate the final magnetic minerals of the burnt clay material. Summary of mineral magnetic parameters measured for Bhon pottery samples are given in Table 1. It is observed that the χ_{LF} values are more evenly spread over a range (24.0126–164.4809)$\cdot10^{-7}$ 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. Most of the samples in the depth of 0–36 cm and 50% of the samples in the depth of 52–59 cm with lower susceptibility indicates the presence of lower concentration of ferrimagnetic mineral. The higher susceptibility values of all samples of other depths show the presence of higher concentration of ferrimagnetic minerals. Samples of all depths show the presence of superparamagnetic grain with size ~0.012–0.023 μm. The BON-3 sample with $\chi_{\mathrm{FD}}\% < 2$ indicates the presence of multidomain particles, but multidomain character is not reflected in further measurement like S-ratio. High values of χ_{FD} indicate the presence of very fine grained metastable magnetic grains spanning the SP-stable single domain (SSD) boundary [13, 14]. All the samples show $\chi_{\mathrm{FD}}\% > 2$ but most of the samples fall in between 4 and 9% suggesting the presence of significant amount of the superparamagnetic magnetite grains.

Dearing et al. [9, 15] have reported that burnt clay samples with $\chi_{\mathrm{FD}}\% > 2$ have detectable concentration of SP grains, and if $\chi_{\mathrm{FD}}\%$ is around ~6–10, samples contain significant amount of fine SP grains of size ~0.012–0.023 μm. Hunt et al. [16] have 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. [15] showed in a model mixing experiment that addition of increasing amount of multidomain-magnetite grains to soil, containing predominantly SP magnetite grains ($\chi_{\mathrm{FD}}\% = 10.5$), causes $\chi_{\mathrm{FD}}\%$ to decrease to <2, while χ_{LF} increases with concentration. Figure 1 shows the χ_{LF} and $\chi_{\mathrm{FD}}\%$ for the samples. The measurement suggests that the pottery samples under investigation are magnetically enhanced materials in terms of concentration and degree of crystallinity of ferrimagnetic mineral magnetite.

3.2. Isothermal remanent magnetization (IRM)

IRM is the remanent magnetization acquired by a sample after exposure to, and removal from, a steady (DC) magnetic field. IRM depends on the strength of the field applied, which is often denoted by a subscript. It is also a function of the magnetic mineralogy and grain size. The maximum remanence that can be produced in a sample is called Saturation Isothermal Remanent Magnetization (SIRM). IRM is often used as an indicator for the presence of ferrimagnetic minerals, but antiferromagnetic minerals, such as hematite and goethite are also capable of acquiring an IRM. After a sample has acquired an IRM it is often possible to (partially) demagnetize the sample by exposing it to a magnetic field of reversed direction. Such a partial

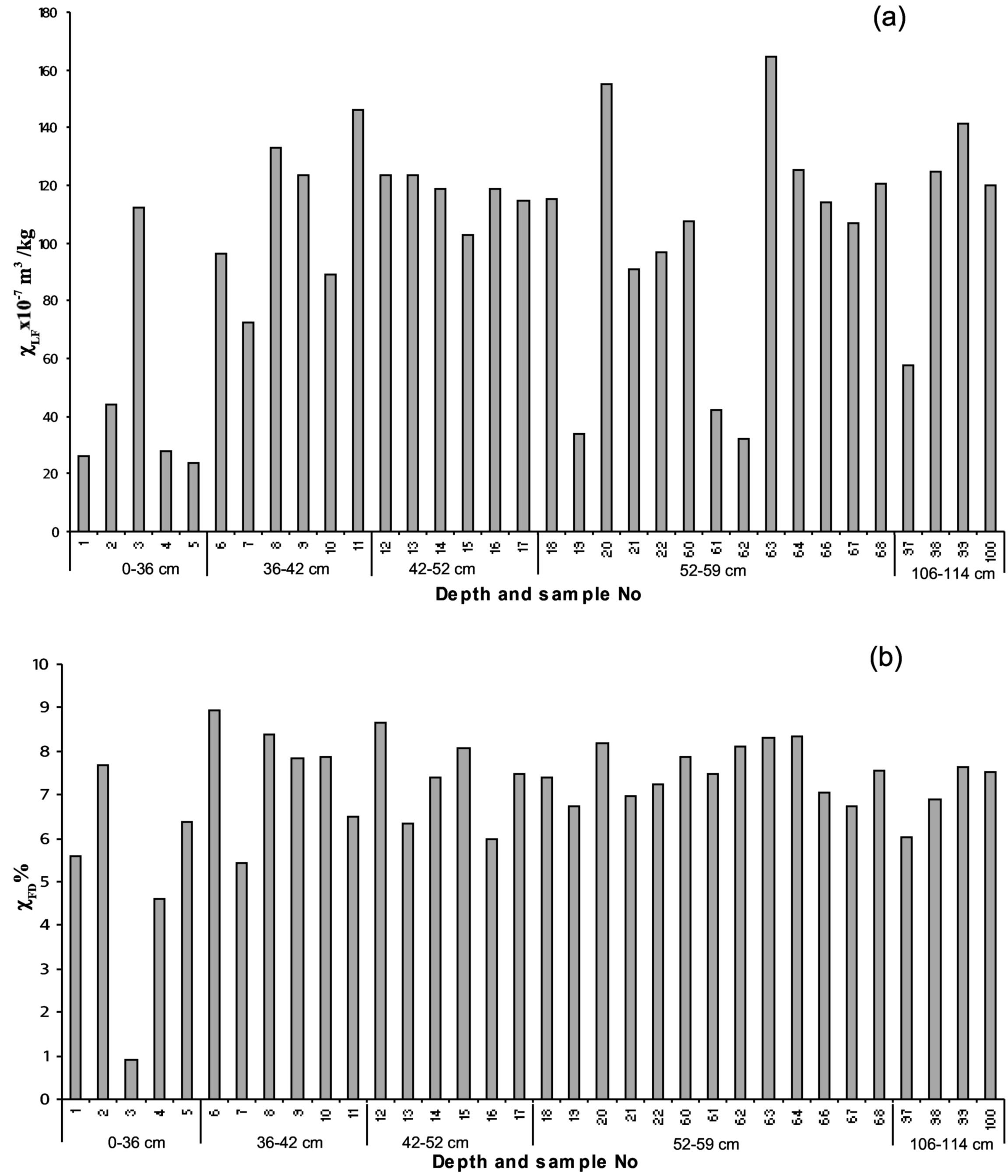

Fig. 1. (a) Low frequency susceptibility (χ_{LF}) and (b) percentage frequency dependent susceptibility (χ_{FD}%) of BON pottery samples.

Table 1. Mineral magnetic parameters of BON pottery samples (n = 34 in total). NRM is natural remanent magnetization, SIRM is saturation isothermal remanent magnetization, χ_{LF} is low frequency susceptibility, $\chi_{\mathrm{FD}}\%$ is percentage frequency dependent susceptibility.

Depth	Sample No	χ_{LF}, 10^{-7} m^3/kg	$\chi_{\mathrm{FD}}\%$	NRM, 10^{-5} A m^2/kg	SIRM, 10^{-2} A m^2/kg	Q-ratio	S-ratio, −300 mT	Soft IRM, 10^{-5} A m^2/kg	Hard IRM, 10^{-5} A m^2/kg
0–36 cm	1	26.0152	5.5737	19.2973	4363.90	1.8642	0.9339	2833.24	288.60
	2	43.7349	7.6728	66.0890	6788.55	3.7978	0.9277	4031.40	490.92
	3	112.0675	0.9142	348.3071	26050.82	7.8110	0.9196	20132.98	2093.76
	4	28.0485	4.6152	29.1345	4770.23	2.6105	0.9248	3394.68	358.70
	5	24.0126	6.3618	7.5552	5453.01	0.7907	0.9267	2507.53	399.70
36–42 cm	6	96.2810	8.9421	21.4482	5402.45	0.5599	0.9092	5108.37	490.56
	7	72.5926	5.4388	20.3133	22944.36	0.7033	0.9516	8338.26	1109.71
	8	133.3333	8.3918	62.2253	12189.58	1.1729	0.9453	8641.96	667.27
	9	123.2769	7.8202	21.5663	14383.28	0.4397	0.9380	9059.45	891.80
	10	88.9283	7.8636	15.8699	12282.47	0.4485	0.9675	6505.93	398.66
	11	146.3615	6.4776	17.9260	21301.01	0.3078	0.9692	14526.40	655.65
42–52 cm	12	123.4007	8.6779	32.5907	6348.82	0.6637	0.9141	5977.47	545.31
	13	123.7595	6.3369	45.8867	17415.26	0.9318	0.9325	9705.64	1175.25
	14	119.0840	7.4081	71.3290	12070.14	1.5054	0.9058	8904.83	1137.10
	15	103.0607	8.0599	17.8211	7028.57	0.4346	0.9042	5505.14	673.24
	16	118.9226	5.9676	54.8050	19459.21	1.1582	0.9743	10362.96	500.85
	17	114.6012	7.4684	74.3037	13854.32	1.6295	0.9132	7674.34	1202.32
52–59 cm	18	115.1009	7.4093	76.3704	11681.21	1.6675	0.9298	7248.45	820.05
	19	33.5788	6.7237	31.0376	5369.99	2.3230	0.9069	3175.37	500.17
	20	155.0933	8.1876	29.7505	15065.18	0.4821	0.9128	9492.64	1314.15
	21	91.0480	6.9544	51.2074	11085.08	1.4135	0.9123	6581.75	972.45
	22	96.5629	7.2598	12.9205	13416.76	0.3363	0.9296	7800.98	944.27
	60	107.3343	7.8712	73.2234	6744.22	1.7145	0.8081	5490.51	1294.18
	61	42.1546	7.4900	24.9226	7664.98	1.4858	0.9384	3526.58	472.30
	62	32.3077	8.1111	31.7195	4820.05	2.4674	0.8905	2971.11	527.63
	63	164.4809	8.3056	162.6944	16299.77	2.4859	0.9267	11406.25	1194.47
	64	125.6039	8.3327	104.9403	1313.96	2.0997	0.9718	10009.66	370.19
	66	113.9535	7.0587	102.0945	15843.35	2.2516	0.9343	8430.13	1041.14
	67	106.7138	6.7318	79.4337	16765.14	1.8707	0.9348	9189.15	1092.57
	68	120.6593	7.5716	156.4655	14208.13	3.2590	0.9721	10505.03	396.32
106–114 cm	97	57.6789	6.0305	13.8749	10904.40	0.6046	0.9164	5650.83	911.98
	98	124.6657	6.8755	72.7850	16505.87	1.4673	0.9617	12119.70	631.67
	99	141.4310	7.6469	26.1320	14956.39	0.4644	0.9112	9714.92	1328.49
	100	120.1893	7.5249	103.7004	11585.82	2.1684	0.9029	7000.87	1124.41

demagnetization can yield information about the ease of remanence acquisition, or the coercivity of a sample. The results are expressed as an S-ratio, for example,

$$S_{100} = \mathrm{IRM}_{-100}/\mathrm{SIRM}\,,$$

where IRM_{-100} denotes an IRM acquired in a reverse field of 100 mT after SIRM acquisition. S-ratios can be used to gain information about magnetic mineralogy [17]. 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. In the present study, all the samples show S-ratio values >0.6 which reflect the presence of ferrimagnetic minerals.

It is also worthwhile to examine the magnetic parameter Koenigsberger's-ratio (Q-ratio $= \mathrm{NRM}/\chi_{\mathrm{LF}} \times$ 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 [18]. The high Q-ratio values are characteristic of stable (thermoremanent) origin of NRM while low values ($Q < 1$) are for other nonstable remanence [19]. The Q-ratios provide a relative importance of remanent and induced magnetization, remanence being dominant for $Q > 1$ [20]. Variations in remanent intensity and susceptibility depend on volume content of magnetite. The Q-ratios >1 indicate the presence of single domain / pseudo single domain

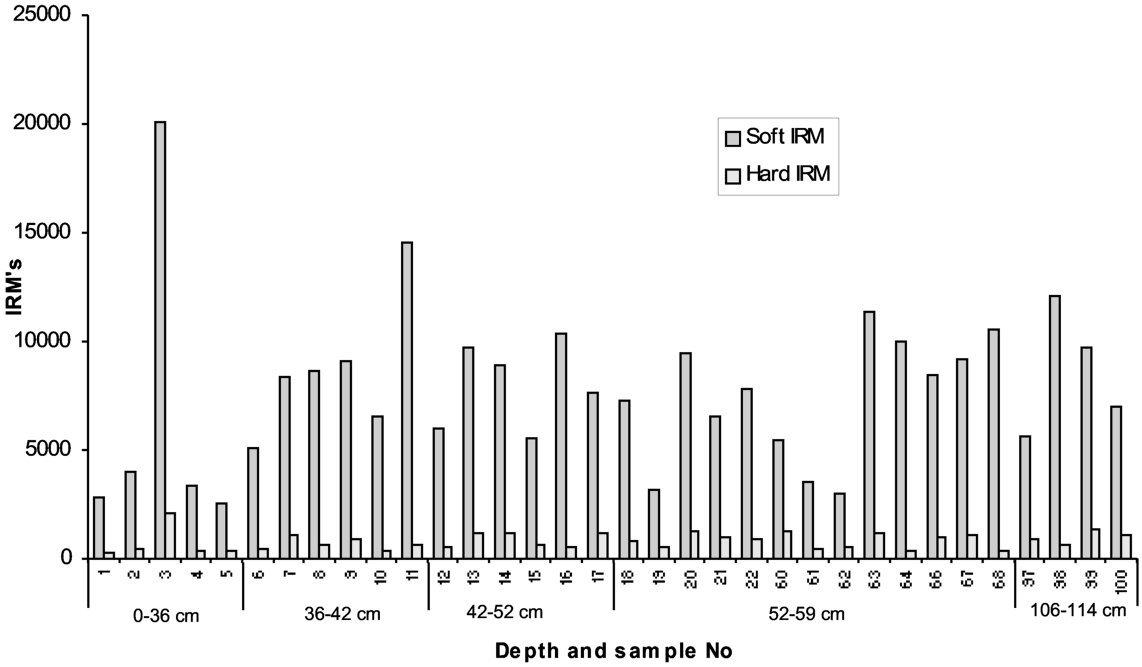

Fig. 2. Isothermal remanent magnetisation of BON pottery samples.

magnetite grains in all the samples and suggest that the samples are suitable for archaeomagnetic analysis. The soft and hard IRM parameters are also indicative of the presence of the type of magnetic minerals. Blomendal et al. [17] have reported that hard IRM is proportional to the concentration of such high-coercivity, antiferromagnetic minerals as goethite and hematite. Basavaiah and Khadkikar [21] have reported that samples having high soft IRM value contain more ferrimagnetic grains than antiferromagnetic grains. In the present study, the higher values of soft IRM point towards the higher concentration of ferrimagnetic mineral.

3.3. IRM acquisition curve

Stepwise saturated isothermal remanent magnetization (SIRM) acquisition curves are useful in identifying magnetic mineral species. SIRM curves up to a maximum field of 1 T and a back-field demagnetization of SIRM are conducted on samples from each flow on a pulse magnetizer (MMPM9). Samples are saturated on applying maximum field of 1 T and they show a remanence coercivity (H_{cr}) of about 40–50 mT, indicating that magnetite is probably the main magnetic carrier. The IRM acquisition curve for three representative samples are given in Fig. 3. In the present investigation, all the samples show remanence coercivity of about 30–40 mT, suggesting that magnetization is carried by low coercivity magnetic mineral such as magnetite/titanomagnetite with low Ti content.

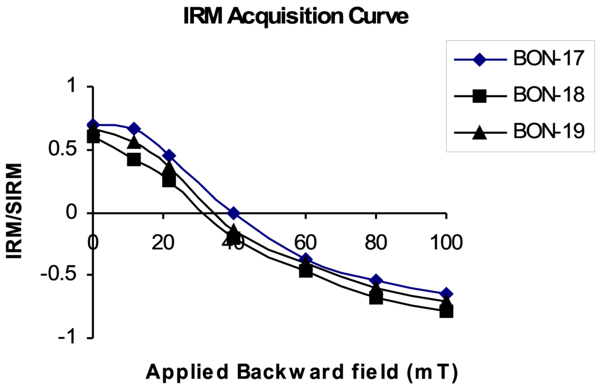

Fig. 3. Isothermal remanent magnetisation acquisition (back-field DC demagnetization curves).

4. Conclusions

1. The obtained higher values of mass specific susceptibility reveals the presence of higher amount of ferrimagnetic minerals from the parent unbaked clay as well as the higher firing temperature achieved during baking.

2. The higher percentage χ_{FD}% values reveal the presence of fine superparamagnetic magnetite particles of varying grain size (∼0.012–0.023 μm).

3. Rock magnetic properties of the pottery samples show stable remanent magnetisation and are found to be suitable for determining paleointensity which is in progress.

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MINERALŲ MAGNETINIŲ PARAMETRŲ PANAUDOJIMAS ARCHEOLOGINIAMS RADINIAMS APIBŪDINTI

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Magnetinės mineralogijos metodais tirtas archeologinių lipdinių šukių rinkinys. Išmatavus izoterminį nuovargį ir jutą silpname lauke, nustatyti tikrieji magnetizmo šaltiniai ir smulkių sudėtinių magnetinių dalelių domeninės būsenos. Visų bandinių magnetinėje mineralinėje sudėtyje dominavo ferimagnetinis mineralas (magnetitas ar magnetitas su nedidele titano priemaiša), tinkamas paleointensyvumui matuoti nustatant senovės geomagnetinio lauko intensyvumą.