

Available online at www.sciencedirect.com



EPSL

Earth and Planetary Science Letters 248 (2006) 508-517

www.elsevier.com/locate/epsl

Reliable absolute palaeointensities independent of magnetic domain state

Mark J. Dekkers ^{a,b,*}, Harald N. Böhnel ^a

^a Centro de Geociencias, UNAM Campus Juriquilla, Queretaro 76230, Mexico ^b Palaeomagnetic Laboratory 'Fort Hoofddijk', Utrecht University, Budapestlaan 17, 3584 CD Utrecht, The Netherlands

> Received 18 January 2006; received in revised form 26 May 2006; accepted 26 May 2006 Available online 17 July 2006 Editor: R.D. van der Hilst

Abstract

Netherlands

Knowledge of palaeointensity variation is required for determining the full vector variation of the geomagnetic field as a function of geological time. This provides essential constraints for numerical geodynamo models. To date, most palaeointensity determination methods are laborious, characterised by rather low success rates, and demand substantial processing time. The rocks under investigation must obey stringent criteria to yield faithful palaeointensities: the magnetic particles must be single domain, the natural remanent magnetisation must be a thermoremanent magnetisation, and during the successive heating steps in the laboratory no chemical alteration should occur. Here, we describe a new method that allows all magnetic domain states to be processed, i.e. it does not require single domain particles. The method makes use of the linearity of partial thermoremanent magnetisation (pTRM) with the applied laboratory field. Multiple specimens are used so that every sample is exposed only once to a laboratory field, warranting that all samples experienced the same magnetic history. Through the limited number of thermal steps alteration effects are reduced as well. The laboratory pTRM and natural remanent magnetisation (NRM) of the specimens are oriented parallel to minimise the effects of high-temperature tails that affect multidomain minerals. The pTRM acquisition temperature is selected below the temperature at which chemical alteration sets in and above the temperature trajectory where secondary viscous NRM components occur. The procedure requires a lower number of steps than any other palaeointensity method, reducing significantly the total time needed per rock unit. We propose to name the new protocol 'multispecimen parallel differential pTRM method'. It provides the correct answer to $\sim 5\%$ for artificial samples and natural rocks containing multidomain magnetic particles that were given a laboratory TRM of known intensity. Application to the Paricutin September–December 1943 lava flow (three sites, 7 specimens per site) yields a weighted mean of $45.9 \pm 1.25 \,\mu\text{T}$, within uncertainty margin of the expected value of $45.0 \,\mu\text{T}$. © 2006 Elsevier B.V. All rights reserved.

Keywords: absolute palaeointensity; magnetic domain state; Paricutin

* Corresponding author. Palaeomagnetic Laboratory 'Fort Hoofddijk', Utrecht University, Budapestlaan 17, 3584 CD Utrecht, The

1. Introduction

The Earth's magnetic field is generated in the liquid outer core of the Earth by magnetohydrodynamic processes referred to as the geodynamo [1]. The intensity of the geomagnetic field as a function of historical and geological time is known to be quite variable. For

E-mail address: dekkers@geo.uu.nl (M.J. Dekkers).

⁰⁰¹²⁻⁸²¹X/\$ - see front matter $\ensuremath{\mathbb{C}}$ 2006 Elsevier B.V. All rights reserved. doi:10.1016/j.epsl.2006.05.040

example, the present-day Earth's magnetic field declines rapidly in strength [2] resulting in a higher exposure to hazardous incoming cosmic radiation because the shielding capacity of the geomagnetic field is reduced. There is little consensus, however, on how far the intensity can drop during periods of stable polarity or what the fieldstrength range is during the reversal process itself [3]. The geodynamo is characterised by two fundamental modes: the so called mixed-polarity status during which the field reverses polarity up to a few times per Ma, alternated by long periods (>20 Ma) during which no polarity changes occur, referred to as superchrons. It is debated whether a relation would exist between reversal frequency and the strength of the geomagnetic field. If so, during superchrons high field intensity would be expected as indeed shown by [4-6]. Other workers [7-10] argue that such relation

The prime reason for these different, occasionally opposing, views is that it is very difficult to acquire goodquality absolute palaeointensity estimates. All studies are based on various versions of the so called Thellier–Thellier technique [11] and involve data selection, usually severe: a large number of determinations must be discarded because they are technically incorrect. For the Thellier–Thellier technique to work the magnectic particles must be single domain and must not alter during the experiment. Moreover, the natural remanent magnetisation (NRM) should be a pure thermoremanent magnetisation (TRM). Though simple, these criteria pose heavy restrictions in practice: most rocks appear to be unsuited.

cannot yet be conclusively discerned.

The thermoremanent magnetisation (TRM) in single domain particles is the sum of partial TRMs (pTRMs) produced between successive temperature segments. Each pTRM segment is independent from one another. In a Thellier-Thellier experiment, the rock's original TRM, i.e. its natural remanent magnetisation (NRM), is progressively replaced by a set of laboratory pTRMs to successively elevated temperatures in a known field. In a 'pTRM gained' versus 'NRM left' plot (referred to as an Arai plot [12]) samples with single domain grains define a straight line from which the palaeofield wherein the TRM was acquired can be calculated straightforwardly, since TRM and therefore pTRM are proportional to the inducing field. Deviations from linearity in the Arai plot are due to multidomain behaviour and/or alteration that preclude a meaningful experiment. Various versions of the Thellier technique have been developed to detect alteration and multidomain behaviour involving zero-field heatings followed by in-field heatings for each temperature step [13], the inverted sequence per temperature step [14], or alternating the zero-field and in-field steps [6]. pTRM checks the test for alteration, while pTRM tail checks [15] and

additivity checks [16] the test for multidomain behaviour that shows up as convex-down in Arai plots. This 'sagging' in Arai plots is due to non-reversibility of the magnetic starting state characteristic of multidomain grains [17,18]. Changing starting states are unavoidable in a Thellier-Thellier experiment. To arrive at more linear Arai plots, a multispecimen approach was proposed recently [19] in which individual specimens are processed for one temperature only; the behaviour for that temperature is evaluated by performing five thermal cycles, including removal of the viscous NRM component (cycle 1 to temperature T_0). Subsequently two zero-field cycles to T_1 and T_2 are performed (with $T_2 > T_1 > T_0$) and thereafter the laboratory pTRM is induced with the field switched on only during cooling from T_2 to T_0 . The final cycle involves demagnetization at T_1 to ascertain the behaviour of laboratory TRM between T_1 and T_0 . All is normalized to T_0 for individual specimens and a composite Arai-style plot allows the determination of the palaeointensity by making use of the inherent within flow variability in magnetic properties. A historic lava flow from Hawaii analysed with this procedure yields the expected intensity within measurement uncertainty [19]. Also recently an entirely new palaeointensity processing method has been developed, based on the ferromagnetic resonance using microwave fields [20,21]. This significantly reduces the thermal alteration and increases the success rate. Microwave palaeointensity determination is also faster than a Thellier type, thermal, experiment. However, due to the complex technology only two laboratories are using it at this moment.

Rather than focussing on optimising the laboratory palaeointensity determination procedure, the alternative approach of searching for rock samples that only contain single domain particles was also adopted, proposing to use submarine basaltic glass [22] to this end, or single plagioclase crystals with magnetite inclusions that are mostly single domain [23,24,4]. The latter are armoured by the surrounding plagioclase [see also [18]] against geological alteration and alteration induced by the laboratory processing. With the exception of the youngest extrusive rocks, weathering induced clay formation is the rule and even seemingly fresh rock samples may bear subtle signs of incipient weathering that only emerge when alteration sets in at temperatures higher than ~450 °C. It should be kept in mind, however, that submarine basaltic glass is relatively rare. Plagioclase crystals are common but the requirement of clear crystals free of groundmass forces the user to small fragments of up to a few mm in size. Meaningful measurement of the accompanying low intensities requires high-resolution superconducting magnetometers whereas the use of standard superconducting magnetometer systems may yield intensities close to their limit of detection that would be difficult to interpret.

2. Rationale of the new methodology

Here, we propose a protocol that is based on the linearity of pTRM with inducing field, a property that is independent of domain state [25-27]. Therefore, it can be applied to all magnetic grains provided no alteration occurs. A further aspect is the consideration of the magnetic history of multidomain particles, i.e. their behaviour during subsequent treatments depends on the previous treatment(s) [17]. So, in a palaeointensity experiment all samples must have exactly the same magnetic history [28,19]. This is achieved by adopting a multispecimen approach in which each sample cq specimen ideally undergoes just one treatment: the acquisition of a laboratory pTRM in a certain field, for all specimens in the experiment to the same temperature. Since the palaeointensity determination is necessarily based on the outcome of several specimens from a lava flow, they should be as identical as possible. A third aspect is the high-temperature tail of multidomain pTRMs [29,17]: by inducing the pTRM parallel to the original TRM vectorial biasing effects are avoided.

If a pTRM is induced parallel to the original TRM in a smaller laboratory field than the palaeofield, the result will be a lower magnetisation than the original TRM. If the pTRM is induced in a stronger field, the result will be a higher magnetisation than the original TRM. If induced in the same field as the palaeofield, no change in remanence will occur. So, the palaeofield is the fieldstrength at which the difference between pTRM and TRM (this is the composite remanence after pTRM acquisition) and the original TRM is zero. Since each specimen contains a different amount of magnetic material, all differences are scaled to the original TRM of the corresponding specimens. We propose to name this protocol 'multispecimen parallel differential pTRM method'. Apart from being independent of magnetic domain state, a substantial additional advantage of this approach is that the temperature of pTRM acquisition can be chosen to be as low as possible to avoid the detrimental effects of alteration. Below we provide the experimental proof in simulated palaeointensity experiments with natural and artificial samples containing multidomain grains and subsequently apply the method to some historic volcanic rocks.

3. Samples and instrumentation

The simulated palaeointensity experiments were carried out on three sample sets labelled ST, PT, and Placer. The ST samples are from the Cretaceous San Telmo pluton, northern Baja California (Mexico) [30]. Its age ranges between 100 and 80 Ma. The samples selected are from sites ST5 and ST6 that show reversible thermomagnetic curves with a Curie temperature of \sim 580 °C and a low coercive force (\ll 10 mT) indicating multidomain magnetite. The PT samples were derived from the Testerazo pluton [31] (sites 12, 13, 15 and 16). The comparatively small plutonic body is considered to be 100-110 Ma in age. Thermomagnetic curves are reversible with a Curie temperature of ~590 °C indicating magnetite while median destructive fields of <10 mT and coercive forces of just a few mT indicate multidomain particles. The Placer sample set is a magnetite-rich (~90% magnetite) heavy mineral concentrate from the Sierra Madre del Sur complex [32] collected from the swash-backswash zone at Manzanillo beach (Mexico). The grain size ranges from $\sim 100 \ \mu m$ to $\sim 600 \ \mu m$ with most grains $\sim 200-300 \,\mu\text{m}$. Most grains are fresh, incipient maghemitisation was noticed on a minor fraction only when examining with reflected light microscopy. Occasionally intergrowths of magnetite and ilmenite were noticed. The concentrate was dispersed into Barro-Mex-Barmelig refractory mortar. After hardening the material was gently crushed with a mortar and pestle and the clumps were set in ZircarTM ceramics alumina cement (#C4002) to obtain smooth sturdy samples able to withstand further handling. Thermomagnetic curves (not shown here) are reversible with a Curie temperature of $\sim 575-580$ °C. Hysteresis loops at room temperature yield a coercive force $(B_{\rm c})$ of 2.66 mT, a remanent coercive force $(B_{\rm cr})$ of 22.98 mT ($B_{cr}/B_{c} = 8.26$) and a ratio of the remanent saturation magnetisation over the saturation magnetisation of 0.020, plotting well within the multidomain field on a Day plot [33]. A first-order-reversal-curve (FORC) diagram [34] shows the dominant vertical contour density at low coercive forces very reminiscent of the FORC diagram acquired by [35] for a 2 mm magnetite crystal. After TRM acquisition, low-field susceptibilities ($*10^{-3}$ SI) ranged from 52.64 to 77.79 (average and standard deviation: 67.305 ± 7.313 , N=22) and TRM intensities (A/m) from 2.89 to 4.775 (average and standard deviation: 3.712 \pm 0.602, N=22). AF demagnetisation of TRM (2 samples) yielded nearly identical demagnetisation curves with median destructive fields of 13.1 and 13.8 mT. The TRMs were virtually completely demagnetised at 100 mT (>98%).

For the TRM and pTRM acquisition experiments an ASC thermal demagnetizer with an in-house built field coil capable of reaching 400 μ T was used. The intensity of the field varied slightly (±2%) along axis of the furnace, during all in-field treatments the samples were placed at the same position in the furnace. Low-field

magnetic susceptibility was measured with an AGICO KLY-3 susceptometer (noise level $2 * 10^{-8}$ SI); remanences with an AGICO JR5A spinner magnetometer (noise level $2 * 10^{-6}$ A/m). AF demagnetisation was carried out with an AGICO LDA-3A alternating field demagnetiser capable of reaching 100 mT peak field; the two-axes tumbler was used. For ARM acquisition a 2 G static alternating field demagnetiser was used (100 μ T DC bias field parallel to alternating field). Thermomagnetic analysis was done in air with classical Curie balances or with a modified horizontal translation balance [36] (heating rate 6 °C, cooling rate 10 °C/min).

4. Simulated palaeointensity experiments

The samples were given a laboratory full TRM by cooling from 650 °C down to room temperature in an (average) field of 50.6 μ T. A classic double heating Thellier–Thellier experiment (Coe version) for samples Placer18, PT16-1z, and ST6-5z shows convex-down Arai plots typical of MD grains, with a lesser curvature for Placer18 (Fig. 1). For the multispecimen parallel differential pTRM method, pTRMs were induced by cooling from 400 °C down to room temperature. Multidomain grains acquire most of their TRM comparatively close to the Curie temperature [37] so for a meaningful difference the pTRM acquisition temperature should be fairly high.

We selected 400 °C because a significant portion of the samples showed erratic behaviour from 500 °C upward during processing of trial multispecimen Thellier-Thellier experiments. These experiments were designed along the lines of [38] and [28] but with the in-field step first as dictated by magnetic history arguments outlined in [28]. Physico-chemical alteration at these comparatively low reheating temperatures was not anticipated because the full TRM was acquired by cooling from 650 °C. In all experiments pTRMs were acquired by having the field switched on during the entire thermal cycle so as to repeat 'nature' as much as possible (the samples are cooled in-field so why reheat them in zero field and switch on just the desired field during cooling?). TRMs and pTRMs were induced in the same furnace so differences in cooling rate potentially biasing the palaeointensity determination [39,40] can be ignored. Also, multidomain titanomagnetite grains may not underestimate the palaeointensity due to rapid cooling as shown experimentally by [41].

In our multispecimen palaeointensity experiments, the low-field susceptibility showed no significant change (1% at most) during the thermal cycling to 400 °C after acquisition of the full TRM. The ST samples underwent a slightly different procedure to mimick palaeomagnetic practice in simulating the elimination of a potentially present viscous remanent magnetisation. This involved thermal demagnetisation at 200 °C before and after the



Fig. 1. Arai plots of simulated palaeointensity experiments of sample Placer 18 (filled diamonds), sample PT16-1Z (filled squares), and sample ST6-5Z (filled triangles). Laboratory 'NRM' — a full TRM by cooling from 650 °C — was induced in a 50.6 μ T field. pTRMs were induced in the same field. NRM and TRM are normalised to their starting and final values respectively calculated with the program Thellier-tool [49]. Temperatures (°C) are indicated along the data points. pTRM checks are indicated by the line segments and open symbols. Placer 18: NRM=4.648 A/m, TRM=4.671 A/m; PT16-1Z: NRM=0.205 A/m, TRM=0.213 A/m; ST6-5Z: NRM=2.196 A/m, TRM=2.373 A/m. pTRM checks at high temperatures are more consistent than those at low temperatures, a manifestation of the non-reproducibility of the starting states characteristic of MD particles. The sagging yields too high palaeointensity values if fitted up to ~500 °C and too low values if the higher temperature segments are being used.



Fig. 2. Multispecimen parallel differential pTRM palaeointensity determinations for an acquisition field of 50.6 μ T. The PT samples are indicated with diamonds, Placer with squares, and ST with triangles. Differences between the TRM and pTRM for each laboratory field are scaled to the TRM of corresponding specimens. The thick line is the least-squares best-fit line, the thin lines are the one-standard deviation confidence zones while the area between the dashed lines represents the 95% confidence zone (two standard deviations). pTRMs were induced by cooling from 400 °C. The ST samples were demagnetised at 200 °C before and after pTRM acquisition to simulate the removal of a viscous component in a 'real' palaeointensity experiment. Actual zero cross-over points of 47.6, 48.4 and 52.7 μ T for Placer, PT, and ST respectively, are close to the expected value of 50.6 μ T, the quality of the line fits as expressed by R^2 is better than 0.965. The different slopes for each sample are the consequence of their slightly varying magnetic domain states.

pTRM acquisition to be able to normalise the pTRM acquired between 200 and 400 °C. Also here susceptibility changes were <1%.

The outcome of the multispecimen parallel differential pTRM protocol is plotted in Fig. 2. Note that we test here whether the multidomain grains show indeed the predicted linear trend. Exactly identical specimens would yield a perfect straight line and the scatter in the observed data is probably caused by subtle differences among the specimens utilised in the experiment. Large multidomain grains acquire most of their TRM close to the Curie temperature and therefore have a smaller pTRM capacity at comparatively low acquisition temperatures than smaller grains that have a more distributed blocking temperature spectrum [37,42]. As expected, all pTRM–TRM differences cross over from negative to positive at nearly the same field value regardless the exact domain state or grain size of their magnetic carriers. The zero-crossing point of the Placer

data is 47.6 μ T (±2.22 μ T, one standard deviation at the zero-crossing point; line fit $R^2 = 0.969$) and that of the PT data 48.4 μ T (±2.23 μ T; line fit R^2 =0.966), just a few μ T below the expected value of $50.6 \,\mu\text{T}$, an encouraging result seen in the perspective that these samples cannot be processed at all with classical Thellier-Thellier protocols. The steeper slope of the PT samples indicates slightly smaller multidomain particles than in the Placer samples. The outcome of the multispecimen ST experiment is plotted in Fig. 2 as well, showing a zero-crossing point at 52.7 μ T (±2.06 μ T; line fit $R^2 = 0.972$, expected value 50.6μ T). Comparison between room temperature values (so before thermal demagnetisations at 200 °C which is warranted here because no viscous remanence is present) would give a zero-crossing point of 48.0 µT (line fit $R^2 = 0.975$). The higher value for the simulated palaeofield with the partial thermal demagnetisation included is likely due to a slightly changed magnetic starting state before the pTRM acquisition. This would result in a higher palaeofield because it is more difficult to change a partially thermally demagnetised magnetic state than a TRM without thermal demagnetisation. The large multidomain ST particles are most prone to show this behaviour. Despite this violation of the ideal experimental procedure deviations are still within 5% of the expected value, a remarkable result.

The importance of using the multispecimen protocol can be appreciated from Fig. 3. Two Placer samples were given incrementing pTRMs, starting at zero acquisition field and subsequently increasing to $100 \,\mu\text{T}$ in $10 \,\mu\text{T}$ steps. Two other Placer samples were given a pTRM starting at a field of $100 \,\mu\text{T}$, and subsequently this field was decreased in $10 \,\mu\text{T}$ steps down to $0 \,\mu\text{T}$. As anticipated, starting in



Fig. 3. Simulated palaeointensity experiments according to the parallel differential pTRM approach but now are multiple fields applied to single specimens of Placer material. Approaching the palaeofield ($50.6 \,\mu$ T) from below (filled symbols) yields too high values whereas approaching from above (open symbols) yields too low values. Differences in slope are the consequences of slightly variable domain states in the samples.

low-fields yielded too high zero-crossing points while starting in high fields yielded too low zero-crossing points. They appear to be not symmetrically distributed around the true zero-crossing point, however, which makes approaching the palaeofield from two sides, by using just two samples, impractical.

5. Palaeointensity of historic lava flows

As a final test the multispecimen parallel differential pTRM protocol was applied to two Mexican historic lavas that have been studied recently, the Paricutin 1943-1952 eruption [43,44] and the Jorullo 1759–1774 eruption [45]. Both volcanoes are monogenetic cinder cones belonging to the Michoacán-Guanajuato volcanic field (Central Mexico). In Paricutin, the NRM of the samples processible with the classical Coe version of the Thellier-Thellier protocol is mostly residing in pseudo single domain titanium-poor titanomagnetite (Curie temperatures ranging from ~450 to ~530 °C) resulting from oxyexsolution that occurred during initial cooling of the flows [44]. The Paricutin palaeointensity [44] yielded good quality but widely varying values for individual samples from 3.8 to 67.5 μ T with an average (±standard deviation) of 43.8± 19.8 μ T for accepted values (N=8) and 35.7±19.6 μ T for all determinations (N=23). The expected palaeointensity is $\sim 45.0 \ \mu T$ based on magnetic observatory data [44] from the Mexico City geomagnetic observatory situated ~250 km East of Paricutin, and on the International Geomagnetic Reference Field models for 1940-1950 [44]. The early stage Jorullo flow was emplaced between 1759-1974 and studied by Gratton et al. [45]. It is a hyperstene normative primitive basalt containing mostly titanium-poor magnetite (Curie temperature between 500 and 540 °C) of pseudo single domain size (median destructive fields of 30-40 mT). Palaeointensity was determined by the Coe variant of the classical Thellier-Thellier technique and by two microwave methods (the 'double heating' and 'perpendicular' methods). Microwave palaeointensities yielded $43.2\pm3.5 \ \mu T$ (N=10, 5 double heating, 5 perpendicular), and Thellier-Thellier palaeointensities gave 53.5±9.2 μ T (N=5); the grand average is 46.6± 6.3 μ T (N=15) [45]. The expected value for the palaeointensity ranges 43–47 µT based on historical field models GUFM1 [46] and CAL37K [47]. In these aforementioned studies of the Mexican historic lavas cooling rate effects were not considered (apparently they are not large since both studies, in particular [45] yielded expected palaeointensities). We did not correct for it as well because the constituting grains are not single domain.

For the present study Paricutin cores were drilled as parallel to the known NRM as practically possible be-

cause the field coil dictates a pTRM direction parallel to the cylinder axis of the specimens (z-component). The three sites (UTM coordinates site 1: 13Q0791607, 2160432; site 2: 13Q0790700, 2161047; and site 3: 13Q0791607, 2160426) comprise a few square meters aerial extent and six cores were drilled each yielding 3 to 4 specimens. Actual inclinations in specimen coordinates varied between 90 and 67°, so the pTRMs were multiplied with the z-component of the TRM divided by the full TRM to take angular differences between pTRM and TRM into account. Although this is not entirely correct for large multidomain grains because of incomplete pTRM additivity, for the present pseudo single domain grains and the relatively small angles the error made is taken to be small. The Jorullo specimens were taken from an existing collection and angles between the pTRM and NRM of up 45° had to be accepted.

The magnetic mineralogy of the three Paricutin sites studied here (all from the September to December 1943 flow), appears to show variable titanium contents. Site 1 behaves according to the previous description [44] while the titanomagnetite from sites 2 and 3 is characterised by a much higher titanium content. After thermal demagnetisation at 200 °C, 80–90% of the NRM is still left in the samples from site 1 compatible with a low titanium magnetite phase. In the other two sites only 20–25% of the NRM is left after demagnetisation at 200 °C. Curie balance curves for all sites showed the Curie temperature slightly higher than 300 °C with serious exsolution setting in at higher temperatures evidenced by non-reversible thermomagnetic curves.

Fig. 4a-c shows the results of the multispecimen parallel differential pTRM protocol per site (i.e. three sites with each 7 specimens, for each palaeointensity determination individual specimens came from three to four cores). The pTRMs were acquired at 300 °C and scaled to the NRM of the corresponding specimen at room temperature. The line fit is the thick line (line fit R^2 s for sites 1, 2, and 3 0.811, 0.912, and 0.762 respectively), the 68% and 95% confidence zones (i.e. one and two standard deviations) are shown by the thin and dashed lines respectively. Site 1 yields a palaeointensity of 45.4 ± 2.55 µT $(\pm$ standard deviation at (pTRM-TRM)=0), site 2: 47.2± 1.60 μ T, and site 3: 41.0 \pm 3.32 μ T. Weighted averaging of the three sites gives: $45.9 \pm 1.25 \,\mu$ T. Merging the three sites and treating them as a single entry (hence based on 21 specimens) yields as palaeointensity 44.7 ± 1.32 µT (Fig. 4d, line fit $R^2 = 0.789$). The outcome is statistically indistinguishable from the expected value of 45.0 µT regardless of the approach adopted. Note that not a single specimen was discarded from the analysis. The curves are somewhat noisy, however, hinting at incipient alteration and/or variations in rock magnetic properties, which — as anticipated — becomes more apparent in the diagram combining all three sites. This could be an explanation for the large variability of the palaeointensity results using the classical approach [44].

In an attempt to reduce the scatter in the data (Fig. 4a–d), initial thermal demagnetisation at 200 °C of

the NRM was carried out with another set of specimens, then pTRMs were induced at 300 °C and subsequently demagnetised at 200 °C. The idea behind the partial demagnetisation is that small neoformed grains that will acquire a pTRM by cooling from 300 °C in a field, are demagnetised again at 200 °C if sufficiently small. Site 1 with 80–90% of the NRM left at 200 °C distinctly



Fig. 4. Application of the multispecimen parallel differential pTRM protocol to historic lavas from the Paricutin (top row, three panels) and Jorullo volcanoes (lower row, right panel). Top row left panel, Paricutin site 1: filled diamonds; central panel, Paricutin site 2: filled squares; right panel, Paricutin site 3: filled triangles. The line fits are for individual sites are indicated; site 1 yields a palaeointensity of $45.3 \,\mu$ T, site 2 of $47.2 \,\mu$ T and site 3 of $41.0 \,\mu$ T (R^2 =0.811, 0.912, and 0.762 for sites 1, 2, and 3 respectively), all very close to the expected value of $45.0 \,\mu$ T. The one-standard deviation (68%) confidence zone is indicated by the thin curved lines, the two-standard deviation (95%) confidence zone is indicated by the dashed lines. The lower row left panel (filled squares) shows the line fit when all data points from Paricutin are grouped together yielding a palaeointensity of $44.7 \,\mu$ T (R^2 =0.789). Confidence zones are indicated the same as for the three top row panels. The central panel (lower row) shows the experiment for sites 1, 2, and 3 with demagnetisation at 200 °C before and after pTRM acquisition (hence another set of samples was utilised). Sites 2 and 3 (open symbols) yield non-interpretable data. For site 1 (filled symbols) the fit improves considerably although the palaeointensity (crossover 45.5 μ T, R^2 =0.896) does not change significantly compared to the previous experiment (top row, left panel). Confidence zones are the same as in the other panels. The lower row, right panel displays the Jorullo volcano data. Open symbols are without thermal demagnetisation at 200 °C before and after pTRM acquisition, the line with the confidence zones corresponds to the fit to the latter data points. Palaeointensity=45.0 μ T (R^2 =0.823).

improved now yielding $45.5\pm2.14 \ \mu T$ (Fig. 4e, line fit $R^2 = 0.896$). Sites 2 and 3 with only 20-30% of the NRM left at 200 °C produced inadequate data because of the unmixing of high-Ti titanomagnetite that had proceeded to such an extent that meaningful palaeointensity data could not be obtained anymore. It is presently unknown why the palaeointensity experiment without previous thermal demagnetisation yields remarkably correct data. The combined effect of heating to 200, 300, and 200 °C again could have (much) more detrimental effects than a single heating to 300 °C, but this remains speculative at present (see also [38] concerning physico-chemical alteration and cumulative annealing time). Anyhow, pTRM-TRM lines with several zero-crossing points as shown in Fig. 4e must be rejected for palaeointensity determination.

The Jurollo specimens (Fig. 4f, 7 specimens constituted one palaeointensity experiment, they derived from 5 drill cores) were processed first only with a pTRM acquisition at 300 °C and in a second experiment (i.e. another set of 7 specimens from five drill cores) with the 200 °C demagnetisation - 300 °C pTRM acquisition - 200 °C demagnetisation sequence as well to suppress the effect of incipient alteration. The results of the first experiment are of low quality (Fig. 4f) presumably because of incipient alteration. Demagnetisation at 200 °C improves the linearity because the altered particles that acquire a pTRM are that small that they are demagnetised by and large at 200 °C. The data are fairly far from perfect yielding $45.0\pm$ 3.27 μ T (line fit $R^2 = 0.823$). This may be related to a large angular difference between the NRM and the pTRM forced upon us, combined with some alteration that could not be corrected for. Nonetheless, the 200 °C demagnetisation data do yield the correct answer of 45.0 µT.

6. Implication and conclusion

The approach outlined in this contribution relies on the temperature selected for pTRM acquisition. Obviously this should be below the temperature where chemical alteration occurs (also classical Thellier– Thellier style experiments cannot be carried out meaningfully when alteration sets in). If there would be a multicomponent NRM its viscous part should evidently be not considered. The temperature of pTRM acquisition must be higher than highest demagnetisation temperature of the viscous component. So, as usual, prior to the palaeointensity experiments, the directional NRM behaviour and the temperature at which substantial alteration would occur must be known. In the present experiments the possible presence of a viscous component was equated to a demagnetisation temperature of 200 °C (the same temperature as in [19]), which has bearing on the magnetic history of the samples. For the large multidomain ST particles the effect of thermal demagnetisation at 200 °C before and after pTRM acquisition amounts to $\sim 4 \ \mu T$ difference in palaeointensity. For pseudo single domain particles the effect is expected to be smaller. Future experiments should determine whether thermal demagnetisation of a viscous component could be replaced by alternating field demagnetisation. As a further check on the reliability of the palaeointensity one could repeat the experiment (with a fresh sample set!) with another temperature of pTRM acquisition. Apart from incipient chemical alteration, a source of variability in the scaled pTRM-TRM(NRM) data points is variability in magnetic properties among the specimens. However, close to the zero-crossing point, the compositional differences become less important: the PT, ST, and Placer lines all go through the same crossing point while they have different slopes.

The present approach differs with existing protocols in that it uses a single temperature and different fields for multiple specimens while others use a single field and different temperatures, in most cases single specimens are cycled through many temperature cycles with alternating field-on and field-off steps. In such cases the magnetic history of non-single domain grains becomes increasingly complex and difficult to predict. In this matter the kinematic modelling by [48] for MD grains that shows increasing sagging in the Arai plot with increasing number of Thellier cycles is noteworthy. Also [6] note that material with vortex hysteresis loops (thus close to true SD grains) would show the most pronounced zigzag behaviour in palaeointensity experiments according to their IZZI protocol, so magnetic history aspects during the experiment may play a pivotal role. If going unnoticed, MD grains with pseudo linear behaviour in Arai plots would yield erroneous, too high palaeointensities. If massive chemical alteration would obscure the second pseudo linear segment at high temperatures, an erroneous (too high) palaeointensity would be deduced from a seemingly technically correct Arai plot.

The procedure proposed by [19] involves few thermal cycles but with two zero-field steps (apart from the unavoidable removal of the viscous component) before the pTRM is given to the specimens (by cooling from 450 °C). The palaeointensity [19] is slightly too high (37 μ T vs. the expected 36 μ T) which might be the effect of non-SD particles in the lava flow. The multispecimen approach of [19] as well as the one proposed here, have in common that the final palaeointensity, even when deduced from only one best-fit curve,

is indeed based on several specimens. Our results include data from at least 7 specimens. Direct comparison with classical methods is somewhat difficult but a flow average for the Paricutin flow based on three sites compares well to that of classsic palaeointensity studies where 2–3 specimens are used. While the multispecimen approach may increase the scatter of data points and reduce the total precision of the obtained results compared to a single sample experiment (but compare the present Paricutin data to the classical approach), based on magnetic history arguments [see also [48]] it is probable that the multispecimen approach retrieves the ancient field intensity more exactly.

The aspect of alteration in situ, the case of a TRM contaminated by geologic processes, is difficult to assess regardless of the palaeointensity procedure adopted. All methods using bulk specimens have to take this possibility into account. Deuteric oxidation at temperatures above the Curie temperature of the magnetic minerals is not important, after oxidation at high temperature the magnetic grains acquire a pure TRM while cooling through their blocking temperature spectrum. Potential alteration at temperatures between 200 and 400 °C is more cumbersome and may lead to the acquisition of a thermochemical remanent magnetisation that would bias the paleointensity determination. In case of doubt whether or not such processes may have occurred, volcanological support for proper site selection is recommended to avoid samples inappropriate for palaeointensity determination.

The multispecimen parallel differential pTRM palaeointensity technique as presented here yields answers correct to $\sim 5\%$ in simulated palaeointensity experiments and for historical lava flows. Its advantages are manifold: no domain state requirement, considerably less processing time because no demagnetisation between consecutive pTRM acquisition steps is required, and less influence of detrimental alteration because the temperature of pTRM acquisition can be chosen optimally. Using the multispecimen approach also allows for modifications of the palaeointensity experiment "on the run", according to the results obtained. Indeed, no specific field increment or sequence is required, as every sample is exposed only once to the laboratory field. For precise data one could select small field increments straddling the expected value based on extrapolation from a few more widely spaced data points. It is expected that more accurate data on intensity fluctuations of the geomagnetic field can be gathered in the future. This would contribute in answering important geophysical questions on the generation and sustainment of the Earth's magnetic field, core-mantle coupling, and inner core growth and formation.

Acknowledgements

This research is a part of the research program of the Vening-Meinesz research School of Geodynamics, Utrecht University. Discussions with F. Martín-Hernández and T.A.T. Mullender is acknowledged. This research was partly funded by the UNAM project IN119202 and DFG project NO334/1.

References

- R.T. Merrill, M.E. McElhinny, P.L. McFadden, The magnetic field of the Earth — palaeomagnetism, the core, and the deep mantle, Acad. Press Geophys. Ser 63 (1996) (531 pp.).
- [2] G. Hulot, C. Eymin, B. Langlais, M. Mandea, N. Olsen, Smallscale structure of the geodynamo inferred from Oersted and Magsat satellite data, Nature 416 (2002) 620–623.
- [3] J.-P. Valet, Time variations in geomagnetic intensity, Rev. Geophys. 41 (2003), doi:10.1029/2002GC000343.
- [4] J.A. Tarduno, R.D. Cottrel, A.V. Smirnov, High geomagnetic intensity during the mid-Cretaceous from Thellier analyses of single plagioclase crystals, Science 291 (5509) (2001) 1779–1783.
- [5] J.A. Tarduno, R.D. Cottrell, A.V. Smirnov, The Cretaceous superchron geodynamo: observations near the tangent cylinder, Proc. Natl. Acad. Sci. 99 (2002) 14.020–14.025, doi:10.1073/pnas.222373499.
- [6] L. Tauxe, H. Staudigel, Strength of the geomagnetic field in the Cretaceous normal superchron: new data from submarine basaltic glass of the Troodos ophiolite, Geochem. Geophys. Geosyst. 5 (2) (2004) Q02H06, doi:10.1029/2003GC000635.
- [7] M. Prévot, M.E.M. Derder, M. McWilliams, J. Thompson, Intensity of the Earth's magnetic field: evidence for a Mesozoic dipole low, Earth Planet. Sci. Lett. 97 (1990) 129–139.
- [8] T. Juarez, L. Tauxe, J.S. Gee, T. Pick, The intensity of the Earth's magnetic field over the past 160 million years, Nature 394 (1998) 878–881.
- [9] P. Selkin, L. Tauxe, Long-term variations in palaeointensity, Philos. Trans. R. Soc. Lond. 358 (2000) 1065–1088.
- [10] A.J. Biggin, D.N. Thomas, Analysis of long-term variations in the geomagnetic poloidal field intensity and evaluation of their relationship with global geodynamics, Geophys. J. Int. 152 (2003) 392–415.
- [11] E. Thellier, O. Thellier, Sur l'intensité du champ magnétique terrestre dans le passé historique et géologique, Ann. Géophys. 15 (1959) 285–376.
- [12] T. Nagata, Y. Arai, K. Momose, Secular variation of the geomagnetic total force during the last 5000 years, J. Geophys. Res. 68 (1963) 5277–5281.
- [13] R.S. Coe, Palaeointensity of the Earth's magnetic field determined from Tertairy and Quaternary rocks, J. Geophys. Res. 72 (1967) 3247–3262.
- [14] M.J. Aitken, A.L. Allsop, G.D. Bussell, M.B. Winter, Determination of the intensity of the Earth's magnetic field during archeological times: reliability of the Thellier technique, Rev. Geophys. 26 (1988) 3–12.
- [15] P. Riisager, J. Riisager, Detecting multidomain magnetic grains in Thellier palaeointensity experiments, Phys. Earth Planet. Inter. 125 (2001) 111–117.
- [16] D. Krása, C. Heunemann, R. Leonhardt, N. Petersen, Experimental procedure to detect multidomain remanence during Thellier– Thellier experiments, Phys. Chem. Earth 28 (2003) 681–687.

- [17] S. Xu, D.J. Dunlop, Thellier palaeointensity theory and experiments for multidomain grains, J. Geophys. Res. 109 (2004) B07103, doi:10.1029/2004JB003024.
- [18] D.J. Dunlop, B. Zhang, Ö. Özdemir, Linear and nonlinear Thellier palaeointensity behavior of natural minerals, J. Geophys. Res. 110 (2005) B01103, doi:10.1029/2004JB003095.
- [19] K.A. Hoffmann, A.J. Biggin, A rapid multi-sample approach to the determination of absolute palaeointensity, J. Geophys. Res. 110 (2005) B12108, doi:10.1029/2005JB003646.
- [20] D. Walton, S. Snape, T.C. Rolph, J. Shaw, J. Share, Application of ferrimagnetic resonance heating to palaeointensity determinations, Phys. Earth Planet. Inter. 94 (1996) 183–186.
- [21] M.J. Hill, M.N. Gratton, J. Shaw, A comparison of thermal and microwave palaeomagnetic techniques using lava containing laboratory induced remanence, Geophys. J. Int. 151 (2002) 157–163.
- [22] T. Pick, L. Tauxe, Geomagnetic palaeointensities during the Creataceous normal superchron measured using submarine basaltic glass, Nature 366 (1993) 238–242.
- [23] R.D. Cotrell, J.A. Tarduno, Geomagnetic paleointensity derived from single plagioclase crystals, Earth Planet. Sci. Lett. 169 (1999) 1–5.
- [24] R.D. Cotrell, J.A. Tarduno, In search of high-fidelity geomagnetic paleointensities: a comparison of single plagioclase crystal and whole rock Thellier–Thellier analysis, J. Geophys. Res. 105 (2000) 23579–23594.
- [25] V.P. Shcherbakov, E. McClelland, V.V. Shcherbakova, A model of multidomain thermoremanent magnetization incorporating temperature-variable domain structure, J. Geophys. Res. 98 (1993) 6201–6216.
- [26] K. Fabian, A theoretical treatment of palaeointensity determination experiments on rocks containing pseudo-single or multi domain magnetic particles, Earth Planet. Sci. Lett. 188 (2001) 45–58.
- [27] K. Fabian, Statistical theory of weak field thermoremanent magnetization in multidomain particle ensembles, Geophys. J. Int. 155 (2003) 479–488.
- [28] A.J. Biggin, H.N. Boehnel, A method to redue the curvature of Arai plots produced during Thellier palaeointensity experiments performed on multidomain grains, Geophys. J. Int. 155 (2003) F13–F19.
- [29] Y. Yu, D.J. Dunlop, On partial thermoremanent magnetization tail checks in Thellier palaeointensity determination, J. Geophys. Res. 108 (2003) B11 2523, doi:10.1029/2003JB002420.
- [30] H. Böhnel, L.A. Delgado-Argote, Palaeomagnetic data from northern Baja California (Mexico): results from the Cretaceous San Telmo batholith, in: H. Delgado-Granes, G. Aguirre-Diaz, J.M. Stock (Eds.), Cenozoic Tectonics and volcanism of Mexico, Boulder Colorado Geological Society of America special Paper, vol. 334, 2000, pp. 157–165.
- [31] H. Böhnel, L.A. Delgado-Argote, D.L. Kimbrough, Discordant palaeomagnetic data for middle-Cretaceous intrusive rocks from northern Baja California: latitude displacement, tilt, or vertical axis rotation? Tectonics 21 (2002) 1049, doi:10.1029/2001TC001298.
- [32] J.F.W. Negendank, N. Nun, Zwei Strandseifen in Mexiko, ihre Zusammensetzung und Herkunft, Geol. Rundsch. 75 (1986) 791–804.

- [33] R. Day, M. Fuller, V.A. Schmidt, Hysteresis properties of titanomagnetites: grain size and compositional dependence, Phys. Earth Planet. Inter. 13 (1977) 260–267.
- [34] C.R. Pike, A.P. Roberts, K.L. Verosub, Characterizing interactions in fine magnetic particles systems using first order reversal curves, J. Appl. Phys. 85 (1999) 6660–6667, doi:10.1063/1.370176.
- [35] C.R. Pike, A.P. Roberts, K.L. Verosub, M.J. Dekkers, An investigation of multi-domain hysteresis mechanisms using FORC diagrams, Phys. Earth Planet. Inter. 126 (2001) 13–28.
- [36] T.A.T. Mullender, A.J. van Velzen, M.J. Dekkers, Continuous drift correction and separate identification of ferromagnetic and paramagnetic contributions in thermomagnetic runs, Geophys. J. Int. 114 (1993) 663–672.
- [37] D.J. Dunlop, Ö. Özdemir, Rock Magnetism Fundamentals and Frontiers, Cambridge University Press, Cambridge (UK), 1997 573 pp.
- [38] K.A. Hoffmann, V.L. Constantine, D.L. Morse, Determination of absolute palaeointensity using a multi-specimen procedure, Nature 339 (1989) 295–297.
- [39] M.H. Dodson, E. McClell, Brown, magnetic blocking temperatures of single domain grains during slow cooling, J. Geophys. Res. 85 (1980) 2625–2637.
- [40] S.L. Halgedahl, R. Day, M. Fuller, The effect of cooling rate on the intensity of weak-field TRM in single-domain magnetite, J. Geophys. Res. 85 (1980) 3690–3698.
- [41] E. McClell, Brown, experiments on TRM intensity dependence on cooling rate, Geophys. Res. Lett. 11 (1984) 205–208.
- [42] D.J. Dunlop, Ö. Özdemir, Effect of grain size and domain state on thermal demagnetization tails, Geophys. Res. Lett. 27 (2000) 1311–1314.
- [43] S. Gonzales, G. Sherwood, H. Böhnel, E. Schnepp, Palaeosecular variation in Central Mexico over the last 30,000 years: the record from lavas, Geophys. J. Int. 130 (1997) 201–219.
- [44] J. Urrutia-Fucugauchi, L.M. Alva-Valdivia, A. Goguitchaichvili, M.L. Rivas, J. Morales, Palaeomagnetic rock-magnetic and microscopy studies of historic lava flows from the Paricutin volcano, Mexico: implications for the deflection of palaeomagnetic directions, Geophys. J. Int. 156 (2004) 431–442.
- [45] M.N. Gratton, A. Goguitchaichvili, G. Conte, J. Shaw, J. Urrutia-Fucugaucho, Microwave palaeointensity study of the Jorullo volcano (Central Mexico), Geophys. J. Int. 161 (2005) 627–634.
- [46] A. Jackson, A.R.T. Jonkers, M.R. Walter, Four centuries of geomagnetic secular variation from historical records, Philos. Trans. R. Soc. Lond. A 358 (2000) 957–990.
- [47] M. Korte, C. Constable, Continuous geomagnetic field models for the past 7 millenia: CAL37K, Geochem. Geophys. Geosyst. 6 (2005), doi:10.1029/2004GC0008000.
- [48] A.J. Biggin, First-order symmetry of weak-field partial thermoremanence in multi-domain (MD) ferromagnetic grains: 2. Implications for Thellier-type palaeointensity determination, Earth Planet. Sci. Lett. 245 (2006) 454–470.
- [49] R. Leonhardt, C. Heunemann, D. Krása, Analyzing absolute paleointensity determinations: acceptance criteria and the software ThellierTool4.0, Geochem. Geophys. Geosyst. 5 (2004) Q12016, doi:10.1029/2004GC00(0807.