A method to reduce the curvature of Arai plots produced during Thellier palaeointensity experiments performed on multidomain grains

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Summary

We highlight benefits resulting from a simple modification to the conventional Thellier–Coe (CTC) method of palaeointensity determination. A consideration of current theory pertaining to partial thermoremanence (pTRM) acquisition in pseudo-single (PSD) and multidomain (MD) grains is used to show that the reversed Thellier–Coe (RTC) method is expected to reduce the curvature of pTRM-NRM plots. Experimental data confirms that this is actually the case although there are some observed effects that are not accounted for by present theory. The RTC method is shown to be an improvement over the CTC method in the sense that it acts to reduce the palaeointensity overestimate produced when an experiment is abandoned below the Curie temperature because of alteration occurring. However, more work is necessary to verify its general applicability.

Key words: magnetic domain, palaeointensity.

1 Introduction

Determinations of the ancient intensity of the geomagnetic field are vitally important in constraining geodynamo models (Dormy et al. 2000) and understanding thermodynamic processes occurring within the Earth (e.g. Biggin & Thomas 2003a). The conventional method for acquiring such determinations is that proposed by Thellier & Thellier (1959) and modified by Coe (1967).

This method requires samples to contain only single domain (SD) grains that fully obey Thellier’s laws in order to be wholly successful; Levi (1977) showed that when pseudo-dingle domain (PSD) or multidomain (MD) particles are present, a concave-up rather than linear Arai plot is produced. Thellier experiments are often abandoned at high temperatures due to thermochemical alteration of the samples and on these occasions it is only the top-left portion of the Arai plot that is used. Biggin & Thomas (2003b) have shown that such a practice can have quite disastrous results: producing overestimates of the palaeointensity by up to 65 per cent even when applied to samples that would normally be regarded as suitable for paleointensity analysis.

Igneous rocks normally contain ferromagnetic grains with a spectrum of sizes centred on PSD and are therefore prone to producing palaeointensity overestimates when normal procedure is followed.

The characteristics of MD grains regarding acquisition and removal of partial thermoremanence (pTRM) have undergone a great deal of investigation (e.g. Shcherbakova et al. 2000; Dunlop & Özdemir 1997). Furthermore, a recent number of articles (Shcherbakov & Shcherbakova 2001; Fabian 2001) have attempted to explain which of the MD behavioural characteristics are responsible for producing the concave-up Arai plots.

In this study, we examine the implications of the current MD theory for a specially designed reversed Thellier–Coe (RTC) experiment. We then present the results of such an experiment as a test for the theory and to demonstrate its usefulness for reducing curvature on a Arai plot.

2 Predictions from theory about the reversed Thellier–Coe experiment

The RTC experiment differs from its conventional counter-part (the CTC experiment) only in that, at each temperature stage, the re-magnetization stage is performed before the demagnetization stage. This method has been suggested for use by others previously (Aitken et al. 1988; Valet et al. 1998) although this was in the context of detecting alteration rather than reducing non-ideal effects due to PSD and MD grains.

The sample is first heated up to some temperature \( T_i \) and cooled back to room temperature \( (T_r) \), all in an applied magnetic field, before the total remanence is measured and recorded as REMAG
The entire process is then repeated except that the magnetic field is held at zero throughout. The second measurement will be referred to as DEMAG ($T_i, T_r$).

The Arai plot can then be produced in exactly the same way as for the CTC experiment. The $y$-axis (NRM) position ($1 - \Delta y$) of each point is simply the (normalized) value of DEMAG ($T_i, T_r$) and the $x$-axis (pTRM) position is calculated from the vector subtraction:

$$\Delta x = \text{REMAG}(T_i, T_r) - \text{DEMAG}(T_i, T_r)$$

At first glance, it does not appear that such a minor change to experimental procedure would cause very significant changes to the results. Indeed, for ideal SD particles, whose unblocking temperatures ($T_{ub}$) exactly equals their blocking temperature ($T_b$), we would expect to obtain identical results. However, it will be shown that significant differences are expected and observed when samples contain PSD and MD grains which do not obey Thellier’s laws of thermoremanence.

Fabian (2000) developed a first-order phenomenological model to explain the behaviour of pTRMs in any grain-size assemblage. He later used the model to synthesize results of CTC experiments (Fabian 2001) in good qualitative agreement with empirical data. This model represents the most complete theory of pTRM behaviour in MD grains to date although it is, by the author’s own admission, far from comprehensive and limited in its usefulness for explaining more subtle behavioural aspects.

Individual samples are described in Fabian’s model as a density function on a matrix comprising blocking temperature ($T_b$) on the horizontal and unblocking temperature ($T_{ub}$) on the vertical. An assemblage of ideal SD grains is thus represented by an infinitely thin ridge going straight from the bottom-left to the top-right corner indicating $T_{ub} = T_b$ at all temperatures. In accordance with the results of Dunlop & Ozdemir (2000), PSD and MD assemblages are represented as symmetrical smears centred on this diagonal indicating that $T_{ub}$ is both greater than and less than $T_b$.

Predictions of this model for the CTC experiment performed on PSD-MD samples can be obtained by considering which regions of the $T_b - T_{ub}$ matrix are affected by each remagnetization and demagnetization stage. However, this model is not presently sophisticated enough to make accurate predictions regarding the results of the RTC experiment because the imparting of a pTRM in a sample that already contains a full TRM was not dealt with by Fabian (2000, 2001). His model, first order as it is, would predict that a lower-right portion of the matrix ($T_i > T_{ub}$) unblocks during the heating but that a new pTRM is not acquired in the portion where $T_b > T_i$. I.E. that the REMAG matrices as given in Fig. 1 would appear identical for the CTC and RTC experiments. This implies that a heating/cooling cycle all occurring while the applied field was not zero would...
result in partial thermal demagnetization and this is not a reasonable prediction. We therefore modify Fabian’s model by assuming that when a sample is heated up to $T_c$, any pre-existing remanence with $T_{ab} < T_c$ adapts to the ambient conditions rather than automatically demagnetizing (e.g. if the field is zero it is demagnetized, if the field is anti-parallel and half the intensity of the field that produced the pre-existing TRM, then it alters its sign and becomes half as strong). Empirical evidence presented later will prove that this assumption, in the context of this study at least, is valid.

In practical terms, we adapt Fabian’s model by replacing $T_0$ on the x axis of each matrix with a new factor $T_{ab}$ which reflects only the blocking temperature for the region of the matrix previously affected by a demagnetization stage. Similarly, $T_{ab}$ on the y axis is replaced by $T_{ab}/T_c$ which defines a horizontal line below which any area already containing remanence is given a pTRM (during a REMAG stage) or is demagnetized (during a DEMAG stage).

Fig. 1 shows the predictions of our modified model for early stages of both the CTC and RTC experiments. Note that despite our modifications to the original model, the CTC experiment produces identical behaviour to that outlined by Fabian (2001).

Fig. 1 predicts that the point resulting from the first double heating stage of the RTC experiment should be positioned on the ideal line of the Arai plot as for SD samples, regardless of the domain state. Subsequent points may fall below the ideal line but will present less of a concave-up curve shape than results from the CTC experiment. The degree to which RTC points are displaced from the ideal line depends on the actual $T_{ab}$–$T_c$ relationship matrix for the particular sample and also the difference in temperature between the double-heating stage that is used to determine the point’s position and its immediate predecessor.

Fabian (2001) argued that non-ideal behaviour in a CTC was only a result of the region of the $T_{ab}$–$T_c$ matrix with $T_{ab} > T_c$. However, his first-order model required this matrix to remain static throughout the experiment. Shcherbakov & Shcherbakova (2001) reported an additional cause of the concave-up Arai plot as the fact that a remagnetization stage following a demagnetization stage to the same temperature imparted less pTRM than a remagnetization stage directly following the sample being fully demagnetized. Following the nomenclature of Shcherbakov & Shcherbakova (2001), the measurement made following:

\[ H = \text{OFF} \]  \( \uparrow \)  $T_c$  \( \downarrow \)  $T_\text{c}$  \( \uparrow \)  $T_\text{r}$  \( \downarrow \)  $T_{\text{r}}$  \( (H = \text{ON}) \]  \( \uparrow \)  $T_\text{r}$  \( \downarrow \)  $T_{\text{r}}$

is referred to as a pTRMb and is less than that made after:

\[ H = \text{OFF} \]  \( \uparrow \)  $T_c$  \( \downarrow \)  $T_\text{c}$  \( (H = \text{ON}) \]  \( \uparrow \)  $T_\text{r}$  \( \downarrow \)  $T_{\text{r}}$

which is referred to as a pTRMb.

This is a measured effect that can only be explained in terms of the partial demagnetization stage modifying the $T_0$–$T_{ab}$ relationship of the sample so that it is more resistant to being magnetized over this interval.

Since a pTRMb is imparted in the CTC experiment but not in the RTC experiment, we would expect this observation to cause observed changes between the results of each of them. The remagnetization stage in the RTC experiment is preceded by a demagnetization to a lower temperature rather than the same temperature as in the CTC experiment. We would therefore expect the demagnetization in the RTC experiment to reduce the acquisition of the subsequent pTRM less than in the CTC experiment leading to a further increase (on top of that predicted by our modified Fabian model) in the calculated value of $\Delta x$ for every stage.

The factors described above tell us that, while the values of $\Delta y$ should remain the same for the results of both experiments, we expect those of $\Delta x$ to be higher for the RTC results and thus for the points on the Arai plot to be closer to the ideal line when this method is used. Current theory appears to predict that, in the context of minimizing the concave-up shape produced by PSD and MD grain assemblages, the RTC method is desirable to the CTC method currently in use. The rest of this article focuses on empirical data to test whether this is actually true.

3 EXPERIMENTAL RESULTS

Six samples of igneous rock with rock magnetic properties in the PSD–MD range were selected for the RTC experiment (Table 1). These had already been made resistant to thermochemical alteration by repeated cyclic heating to 700°C. Measurements of remanence were made using an Agico JR-5 magnetometer and heating/cooling cycles were performed in an ASC thermal demagnetizer.

First, as a pre-check of pTRM behaviour, four of the samples were given a full TRM in a field of 50 $\mu$T and were subsequently subject to three consecutive DEMAG (300, $T_c$) stages of heating, cooling, and measurement. These were then given the same full TRM again and were first subject to a REMAG (300, $T_c$) stage in the same applied field before being subject again to four consecutive DEMAG (300, $T_c$) stages.

The results of this experiment are illustrated in Fig. 2 and this allows three important observations to be made. First, each demagnetization stage removes progressively more ‘NRM’ than the previous one although the effect grows less each time. This process removed a total amount of 2–4 per cent of the ‘NRM’ and has no foundation in the theory described above. Control samples with more SD-like properties were treated and measured alongside those described here. These exhibited variations in the DEMAG (300, $T_c$) of less than half a percent indicating that experimental error cannot explain the results.

The second observation is that the measured remanence is essentially identical before and after the REMAG (300, $T_c$) stage which directly follows the imparting of the full TRM. The final important observation that can be made from Fig. 1 is that for all samples, the remanence measured after the first demagnetization stage (that made directly after the full TRM was imparted) is 2–3 per cent less than that measured after the fourth demagnetization stage which was made subsequent to the REMAG (300, $T_c$) stage. The second and third observations were also apparent in the results of the next experiment and shall be discussed below.

Following this experiment, all six samples were given a full TRM in a field of 50 $\mu$T and were subject a CTC experiment using a parallel field of 25 $\mu$T. The entire process was then repeated using the RTC method. The Arai plots produced from these two experiments are shown in Fig. 3.

**Table 1.** Hysteresis parameters for the samples used in this study. The last column provides the high temperature tail of a pTRM imparted between 300°C and $T_c$ (as the sample was cooling from $T_c$) as a percentage of the pTRM itself.

| Sample | $M_{RS}/M_S$ | $H_C$ (mT) | $H_{CR}/H_C$ | Tail[pTRM(300, $T_c$)]
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
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<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.16</td>
<td>16.2</td>
<td>1.93</td>
<td>15.2 per cent</td>
</tr>
<tr>
<td>2</td>
<td>0.11</td>
<td>12.6</td>
<td>1.97</td>
<td>15.9 per cent</td>
</tr>
<tr>
<td>3</td>
<td>0.10</td>
<td>10.9</td>
<td>1.98</td>
<td>14.7 per cent</td>
</tr>
<tr>
<td>4</td>
<td>0.10</td>
<td>11.9</td>
<td>1.98</td>
<td>17.1 per cent</td>
</tr>
<tr>
<td>5</td>
<td>0.09</td>
<td>9.0</td>
<td>2.00</td>
<td>18.9 per cent</td>
</tr>
<tr>
<td>6</td>
<td>0.09</td>
<td>10.5</td>
<td>1.98</td>
<td>19.6 per cent</td>
</tr>
</tbody>
</table>

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The most important results presented by these plots are listed below.

1. Except at high temperatures, all RTC points represent a marked improvement, in terms of proximity to the ideal line, over the CTC points.
2. The first RTC point (300 °C) for all samples falls precisely on the ideal straight line.
3. The majority of RTC points are displaced primarily to the right of, but also up from, their CTC counter-parts.
4. Some points from both the CTC and RTC experiments cross the ideal line and are located marginally above it at high temperatures.

The RTC method therefore does indeed have the potential to be used in palaeointensity investigations as a means of reducing Araki plot curvature.

The prediction of the modified Fabian model was that the second RTC point (that following zero pTRM) would fall on the ideal line whereas the others would fall below it but not to the same degree as the CTC points. This is entirely borne out by the experimental results presented here confirming that it is an excellent first order model and that our assumption regarding the imparting of a pTRM in a sample which already contained a full TRM is entirely valid.

Nevertheless, a few of the smaller details do not agree and these may be of fundamental importance in uncovering a complete picture of MD pTRM theory. These will be discussed in the next section.

Observation number 4 of those given above should be given some attention before proceeding. We believe that the crossing, by points, of the ideal line is likely to be a result of the acquisition of a small self-reversed component close to the Curie temperature. Self-reversal of thermoremanence to varying degrees has been reported previously in MD samples by several authors (McClelland & Sugiura 1987; Sugiura 1988; Shcherbakov et al. 1993; McClelland & Shcherbakov 1995). The self-reversed components have been observed to be imparted close to the Curie temperature of magnetite, in the range 560–580 °C. They were therefore interpreted by McClelland & Shcherbakov (1995) as an expression of interaction between inhomogeneous regions, with this range of Curie temperatures, within the grains of magnetite.

Observation number 4 above, suggests that the self-reversed component was removed by the demagnetization stages at some temperature below 500 °C allowing the points to cross the line, but then was not imparted again until the highest temperature remagnetization stages. This 're-impartment' then brings the points back to the ideal straight line at zero NRM.

An alternative explanation could be that the high temperature points cross the ideal line because they consist of both residual NRM and coexisting tails of acquired pTRM. Although there is little evidence from the present study to discriminate against this alternative, the authors prefer the former explanation for reasons provided in the next section.

4 DISCUSSION

The first of the incongruities between the theory of Fabian (2000) and the empirical evidence was observed in both the CTC–RTC and the initial experiments. It is that a demagnetization stage following a remagnetization stage to the same temperature removes a smaller amount of TRM than a demagnetization stage performed following either a full TRM being imparted or a remagnetization stage to a lower temperature. The most obvious explanation for this is that a remagnetization stage imparts a small high temperature tail that is not removed by a subsequent demagnetization stage to the same temperature. There are however, several reasons to not immediately accept this explanation. One of these reasons is simply that it requires the tail of the recently imparted pTRM to co-exist, over a similar temperature range, with the remaining TRM of the sample. This is not an intuitively pleasing notion, requiring certain
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Figure 3. Arai plots for the six samples studied here (sample number given in top right corner of each plot). Hollow (filled) points are results from the CTC (RTC) experiment. The dashed line is the ideal straight line given for reference. Temperatures for stages are given for sample 1 and are identical for the other samples. Note that there was no 300°C (200°C) step in the conventional (reversed) experiment.

regions within the boxes shown in Fig. 1 to contain NRM and pTRM simultaneously.

Other reasons to reject this explanation are empirically-based. Biggin & Thomas (2003b) performed CTC experiments incorporating pTRM tail checks (Riisager & Riisager 2000) on samples with PSD properties. Despite these samples producing Arai plots that were concave-up to some degree, their tail checks failed to detect any surplus magnetization imparted after the remagnetization stages. Similar experiments performed on crushed magnetite in the MD size range have also failed to detect tails (D.J. Dunlop, personal communication). High temperature pTRM tails are a robust characteristic for pTRMs formed within PSD–MD samples that were previously completely demagnetized. However their role in samples which already contain a pTRM or even a full TRM remains to be elucidated. A forthcoming paper (Biggin & Böhnel, in preparation) will focus on this problem.

The other possible explanation for the observed results lies in the fact that the $T_b - T_{ib}$ relationship for the sample can be modified by the heating/cooling cycles that comprise the experiment. This type of behaviour is presently not represented in the Fabian model but has been shown to exist (Shcherbakov & Shcherbakova 2001).

Several authors have reported that the number of domains within a grain is temperature dependent (e.g. Heider et al. 1988). McClelland & Shcherbakov (1995) showed that the domain configuration of grains could also be different at room temperature depending on whether the sample was previously thermally demagnetized or AF demagnetized. This suggests that the domain configuration is also sensitive to its thermal and magnetic prehistory. We argue, therefore, that thermal cycling involving the same temperatures also acts to modify the domain configuration in MD grains and that this is the cause for the observed changes in the $T_b - T_{ib}$ relationship.

The observation made here is that:

\[ (H = \text{ON}) T_r \uparrow T_c \downarrow T \downarrow T_r (H = \text{OFF}) \uparrow T_r \downarrow T \]

is greater than that made after:

\[ (H = \text{ON}) T_r \uparrow T_c \downarrow T_r (H = \text{OFF}) \uparrow T_r \downarrow T_r \]

This relationship is the \textit{exact} opposite of that given earlier for pTRM$^a$ and pTRM$^b$. In the former instance, the surplus demagnetization stage makes the sample more resistant to remagnetization. Therefore, in this instance it seems feasible that the surplus demagnetization stage makes the sample more resistant to demagnetization. This logical simplicity is part of the reason why the authors favour this explanation over the presence of a high temperature tail.

The only other behaviour observed here that is not accounted for by the described theory is the progressive removal of TRM

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by repeated demagnetization stages involving the same temperatures. This can also be qualitatively explained by assuming that thermal cycling leads to a shift in the domain structure of individual grains (and therefore the $T_b - T_{ub}$ relationship for the sample).

Simply, cyclic heating and cooling to 300 °C allows each grain to modify its domain configuration progressively further towards a demagnetized state. Obviously, we would expect there to be some limit to how demagnetized a sample could become by repeated heating/cooling cycles to 300 °C (viscous effects are unimportant because each cycle lasts for the same amount of time). Fig. 2 shows that the amount of progressive demagnetization that occurs for the four samples decreases with each cycle which suggests that this limit was being approached during this experiment.

This study represents only a preliminary investigation of the RTC method as a means of reducing non-ideal behaviour caused by PSD and MD grains. The results acquired so far are very promising but much follow-up work is required.

First, we have shown that the method works fine if the magnitude of the field used in the experiment differs from that used to impart the full TRM. However, it would also be useful to test its sensitivity to differences in the direction of the applied field. The modified Fabian model, as illustrated in Fig. 1, suggests that the results should be independent of changes in applied field direction. Real palaeointensity experiments seldom use an applied field exactly parallel to the NRM and therefore this should be confirmed empirically.

Equally, although we would not expect the qualitative results to differ in any way if we were to repeat the experiment using samples with entirely different PSD/MD properties, it would be useful to confirm this using samples that exhibit more extreme concave-up behaviour than those described here.

Finally, we will discuss how the RTC method can practically be adopted for use in palaeointensity studies. Secondary magnetizations are a feature encountered in most palaeointensity studies that were not present here. In principle, these should not present any more problems for the RTC method than for the CTC method providing that one ensures that points on the Arai plot that are influenced by them are not used to produce the field estimate. Under any circumstances, a palaeointensity study should always be preceded by a paleodirectional study of sister-samples that uses step-wise demagnetization only.

Finally, thermochemical alteration will play no greater or lesser part in RTC experiments than in their conventional counterparts. Nevertheless, the whole purpose of using the new method is that, should the experiment need to be abandoned because of its onset as usually occurs, the remaining segment will be less prone to producing an overestimate of the palaeointensity. It should be possible to insert pTRM checks into an RTC experiment in much the same way as with the CTC method although this also should first be investigated.

A potential problem with the RTC method is inherent to the fact that, in contrast to the CTC method, a REMAG stage is carried out before the DEMAG stage at a given temperature. If, as might be expected, the first heating to a temperature causes the most alteration and produces a new ferromagnetic phase with high blocking and unblocking temperatures then the RTC method will impart a chemical remanent magnetization (CRM) to this phase whereas the CTC method will not. It should be possible to observe this alteration from the subsequent DEMAG stage but nevertheless, this represents the type of problem which needs to be evaluated before the CTC is replaced.

A detailed follow-up study is currently underway that aims to further investigate the effectiveness of the RTC method and also to investigate other means of straightening concave-up plots.

5 CONCLUSIONS

(1) Both current theory and empirical evidence strongly suggests that the reversed Thellier–Coe method leads to a reduction in the degree of curvature of plots produced by all PSD and MD grains.

(2) This reduction is due primarily to the remagnetization stage now being able to affect parts of the pre-existing TRM before they are demagnetized. This moves the points to the right on the Arai plot.

(3) The points are moved further still to the right because the remagnetization stages are no longer preceded by a demagnetization stage to the same temperature which usually acts to make the sample more resistant to remagnetization (Shcherbakov & Shcherbakova 2001).

(4) The points are also moved slightly up from their conventional counterparts on the Arai plot. We argue that this is not a result of a pTRM tail being imparted but is rather due to the sample being made more resistant to demagnetization by the preceding remagnetization stage.

(5) These samples progressively demagnetize when they are repeatedly heated and cooled in zero field over the same temperature range. This is probably caused by the grains comprising them progressively modifying their domain structure to a more demagnetized state.

(6) The RTC method appears to have enormous potential to reduce non-ideal behaviour related to PSD and MD grains but it stressed that further research is necessary to clarify its general applicability.

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