J. Geomag. Geoelectr., **27**, 245-255, 1975

Comparison of Two Methods of Performing the Thellier Experiment (or, How the Thellier Experiment Should Not Be Done)

Shaul LEVI*

Geophysics Program, University of Washington, Seattle, Washington, U.S.A.

(Received June 6, 1975; Revised August 25, 1975)

Described are two slightly different procedures for performing the Thellier experiment (THELLIER and THELLIER, 1959). The first method is essentially that of COE (1967a, b), and the second is a slight modification thereof. The observed differences in the PNRM-PTRM curves are attributed to differences in the experimental procedure. The Coe method is preferred, as it is more symmetrical with respect to high temperature VRM and spontaneous decay and more likely to yield linear PNRM-PTRM plots.

1. Introduction

The Thellier method for paleointensity determinations depends for its success on the additivity and independence of partial-TRMs, PTRMs, acquired in different temperature intervals (THELLIER, 1938, 1946) and on the assumption that both TRM and NRM (assumed to be TRM) are linearly proportional to the external field, $\boldsymbol{h}$. Thus in the absence of physical and chemical alterations upon heating one obtains:

$$\frac{|\overrightarrow{\mathrm{PNRM}}(T_1, T_R, \boldsymbol{h})|}{|\overrightarrow{\mathrm{PTRM}}(T_1, T_R, \boldsymbol{h}_L)|}=\frac{|\boldsymbol{h}|}{|\boldsymbol{h}_L|}$$

where $\overrightarrow{\mathrm{PNRM}}$ $(T_1, T_R, \boldsymbol{h})$ is the partial-NRM that is lost upon heating the sample from room temperature, T_R, to a higher temperature, T_1. $\boldsymbol{h}$ is the unknown field in which the NRM was acquired. $\overrightarrow{\mathrm{PTRM}}$ $(T_1, T_R, \boldsymbol{h}_L)$ is the partial-TRM produced by cooling the sample from T_1 to T_R in the known laboratory field, $\boldsymbol{h}_L$. The sample is heated to successively higher temperatures until no remanence remains. The data thus generated consists of PNRM$\equiv \Delta J_{NRM}$, PTRM$\equiv \Delta J_{TRM}$ pairs for the different temperature intervals. Ideal behaviour in the Thellier

* Currently at the Department of Geology and Geophysics, University of Minnesota, Minneapolis, Minnesota, U.S.A.

sense implies that for a given sample the same ratio $|\boldsymbol{h}|/|\boldsymbol{h}_L|$ is obtained for each temperature interval. The data can be represented graphically by plotting ΔJ_{NRM} versus ΔJ_{TRM} for the different temperatures (ARAI, 1963, in COE, 1967 b). Ideally behaving data will be linear with slope $-h/h_L$.

COE (1967 b) discussed many of the physical mechanisms that might lead to non-linear PNRM-PTRM plots. In this study two methods for performing the Thellier experiment are compared. Prepared samples containing different species of magnetite particles are used, and both methods are always performed on the same sample. Since all the experimental variables are controlled and the same sample is used, the experiments are designed to accent procedural differences between the two methods for doing the Thellier experiment.

2. Preparation of Samples and Their TRM Properties

The samples studied were prepared by dilutely dispersing magnetite powder (about 1% by weight) in a matrix of high purity alumina and calcium aluminate cement. The samples are molded cylinders about 24 mm in diameter and 22 mm in height. The samples weigh between 15 and 19 g. The samples' magnetic and physical properties are stabilized by heating to 650°C for six hours in a slightly reducing environment, using residual nitrogen gas at about 10^{-1} Torr and carbon as a reducing agent, to prevent oxidation of the magnetite; all subsequent heatings during the Thellier experiments were done in an identical heating environment. Some of the properties of the magnetite powders (previously heated to 650°C in a reducing environment) and the TRM properties of the heated samples are described in Table 1.

In column 1, sample and powder numbers are listed. (Samples 8 and 7 contain different species of magnetite particles.) In column 2, the origin of the magnetite powders and their predominant particle shapes are described. In the third column both mean and maximum particle sizes are listed. Particle sizes and shapes were obtained from electron microscope pictures. Column 4 lists the powders' Curie points. Column 5 gives the samples' TRM acquired in a 0.46 oe field. (The large difference in the TRM intensities of samples 8 and 7 is largely due to the difference in their magnetite powder concentration.) $H_{\frac{1}{2}}$ is the median demagnetizing field, and $T_{\frac{1}{2}}$ is the median demagnetizing temperature, defined as the peak alternating field or temperature required to demagnetize the TRM to half its original value. In column 8 J/J_{TRM} gives the samples' TRM decay after each of three successive cooling cycles in zero field to 78°K and then reheating to T_R. Remanences are always measured at T_R. The data of Table 1 show that samples 8, 7 and 2 are representative of samples frequently encountered in paleomagnetic studies of igneous bodies.

Table 1. Physical Characteristics of the Magnetites and TRM Properties of the Samples.

Sample (powder) No.	Particle origin and shape	Particle size d, μm	T_C, °C ±10°C	TRM (×10^{-3} gauss) h=0.46 oe	$H_{1/2}$, oe	$T_{1/2}$, °C	J/J_{TRM} liquid N_2 cycles in h=0
8	Synthetic	$\bar{d}=0.12\pm0.04$	575	1.08	292±10	400±15	0.990
	Regular polyg.	$d<0.3$					0.970
	Spheres to cubes						0.965
7	Synthetic	$\bar{d}=0.21\pm0.06$	570	13.2	360±15	508±10	0.965
	Regular polyg.	$d<0.5$					0.961
	Spheres to cubes						0.955
2	Natural	$\bar{d}=2.7$	580	0.227	78±4	475±15	0.590
	Crushed, sieved	$d<150$					0.465
	Irregular						0.476

1 oersted=10^{-4} w/m^2=10^{-4} tesla
1 gauss=1 emu/cm^3=10^3 amp/m

Samples 8 and 7 contain synthetic, submicron magnetite particles and their magnetization is quite stable; the remanence probably resides in single domain or pseudo-single domain particles. A substantial fraction of the remanence of sample 2 resides in multidomain particles; this is supported by its larger particle sizes, its lower TRM stabilities with respect to alternating fields, and the large fraction of its remanence decaying after low-temperature cycles in zero field.

3. Experimental Procedure for the Thellier Experiments

The first sequence of the Thellier experiments was executed following COE (1967a, b).

a. The NRM (which is a laboratory TRM) is measured at room temperature, T_R.
b. The sample is heated to $T_1 > T_R$ and cooled back to T_R in zero field; the magnetization is measured at T_R to obtain the PNRM lost between T_R and T_1.
c. The sample is reheated to $T_1 > T_R$ and cooled back to T_R in the presence of the external field, $\boldsymbol{h}_L$. *The field* $\boldsymbol{h}_L$ *is continuously present throughout the entire heating and cooling cycle.* The magnetization is measured at T_R to obtain the PTRM acquired between T_1 and T_R.

Steps b and c are repeated at successively higher temperatures until all the blocking temperatures are exceeded. The temperature is maintained at a particular elevated temperature to allow thermal equilibrium to be established. Depending on the temperature, the time at T_1 varies from 30 to 75 minutes. (Longer times at T_1 are used for progressively higher temperatures.)

The second sequence of Thellier experiments is identical to the first with one exception: in step c the sample is reheated to $T_1 > T_R$ in zero field, and the external field, $\boldsymbol{h}_L$, is turned on about 5 minutes prior to cooling. (Chronologically, the second sequence was executed first, under the belief that this procedure would prevent the introduction of high temperature VRM (viscous remanent magnetization).)

The laboratory field, $\boldsymbol{h}_L$, in these experiments is 0.460 ± 0.002 oe. All the remanences (including the NRM) are laboratory produced TRMs or PTRMs and are always along the axes of the sample cylinders. The field during step b is nulled to within $\pm 50\gamma$ in the region of the experiment. The reproducibility of the temperature during a particular step of the paired heatings is determined by an automatic temperature controller and is thought to be better than $\pm 3\,^\circ$C.

4. Results and Discussion

The PNRM-PTRM curves of samples 8, 7 and 2 are shown in Figs. 1–3.

The magnetization is always normalized with respect to the NRM. The numbers associated with the (PNRM, PTRM) points correspond to the temperature in °C. The initial point is defined as (1.000, 0.000) and small deviations of the final points from (0.00, 1.00) are attributed to small changes of the samples' TRM upon heating. The lines are drawn connecting the initial and final points and they represent "ideal" behaviour in the Thellier sense. The two sets of data in each figure are obtained using the same sample. The upper data (squares)

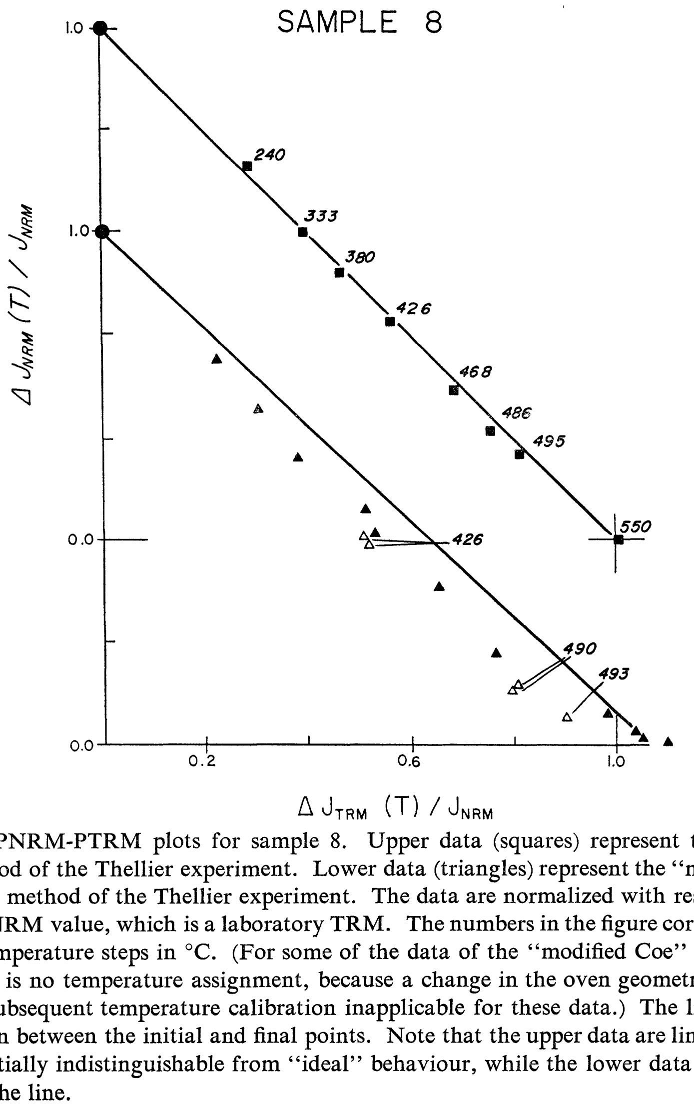

Fig. 1. PNRM-PTRM plots for sample 8. Upper data (squares) represent the Coe method of the Thellier experiment. Lower data (triangles) represent the "modified Coe" method of the Thellier experiment. The data are normalized with respect to the NRM value, which is a laboratory TRM. The numbers in the figure correspond to temperature steps in °C. (For some of the data of the "modified Coe" method there is no temperature assignment, because a change in the oven geometry made the subsequent temperature calibration inapplicable for these data.) The lines are drawn between the initial and final points. Note that the upper data are linear and essentially indistinguishable from "ideal" behaviour, while the lower data sag below the line.

$J_{TRM} = 0.71 \times 10^{-3}$ emu/g for $h_L = 0.46$ oe.

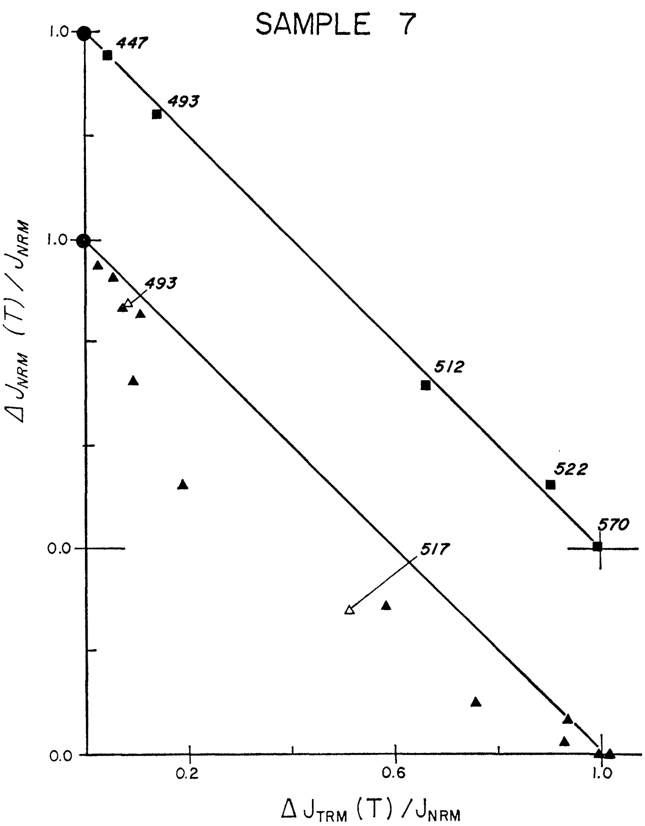

Fig. 2. PNRM-PTRM plots for sample 7. Figure format is identical to that of Fig. 1. Note that the upper data are linear and essentially indistinguishable from "ideal" behaviour, while the lower data sag below the line.
$J_{TRM}=7.7\times 10^{-3}$ emu/g for $h_L=0.46$ oe.

were obtained by the Coe method of the Thellier experiment, and the lower data (triangles) were obtained by "modified Coe" procedure. Each open triangle represents a repeat experiment conducted after the completion of both continuous methods of the Thellier experiment to test the reproducibility of the data of the "modified Coe" method. Each open triangle represents a single temperature "modified Coe" experiment, where after inducing a total TRM in the sample, only one PNRM, PTRM pair is obtained at a single temperature, whereupon the sample is given another total TRM.

The upper data obtained by the Coe method yield linear PNRM-PTRM plots for samples 8 and 7. The slope of the line obtained from the least squares fit of all the points is -1.02 for sample 8 and -0.98 for sample 7, whereas the ratios of initial to final points are -1.00 for both samples. Thus for samples 8 and 7 the Coe method of the Thellier experiment produces ideal behaviour in the Thellier sense.

The lower data obtained by the "modified Coe" method sag below the line connecting the initial and final points and are concave up. A paleointensity determination using a linear approximation to the lower temperature points would yield slopes that are substantially greater than unity, leading to apparent paleointensities that are anomalously high. This is illustrated by the linear least squares fit of the lower temperature data, covering at least half the samples' NRM, and yielding paleointensities that are high by 9% and 21% for samples 8 and 7, respectively. On the other hand, a linear approximation to the higher temperature points leads to apparent paleointensities that are too low.

The data of the "modified Coe" procedure for sample 8 provide a particularly instructive example. The ratio of the final to initial points, TRM/NRM= −0.90, yields an intensity which is 10% lower than h_L. This result is due to some reduction of the magnetic minerals, indicating that the sample's magnetic properties were not fully stabilized when the continuous "modified Coe" experiment was executed. (It should be recalled that, chronologically, the "modified Coe" experiment was done first.) In contrast, the slope of the line through the lower temperature points, but including at least one-half of the sample's NRM, yields an apparent intensity that is 9% greater than h_L. Thus it appears that the effect of the "sag" is actually greater than 9% and closer to 19%. This statement is supported by the single point "modified Coe" experiments, which were obtained after both the continuous "modified Coe" and the Coe methods of the Thellier experiment were completed, when the sample's magnetic properties were more fully stabilized. It is seen that the single point "modified Coe" experiments for all three samples substantiate the "sag" of the continuous "modified Coe" experiments. However, for a given temperature, the open triangles for sample 8 consistently sag below the closed triangles.

It appears that while the "modified Coe" method carefully avoids high temperature VRM, it introduces excessive spontaneous decay of the remanence. In the Coe method the time at elevated temperature during the thermal demagnetization step is balanced by equal time at the elevated temperature under the influence of an external field during the PTRM steps, such that the spontaneous decay is balanced by the high temperature VRM. For the "modified Coe" procedure the samples spend about double the time at the elevated temperature in zero field, the external field being turned on only for the last 5 minutes of the PTRM heating (step c) thereby increasing the spontaneous decay with respect to the high temperature VRM. This is consistent with the data of Figs. 1 and 2 where for a given temperature the PNRM lost is greater than the PTRM gained. It is expected that the deviation of the data obtained by the "modified

Coe" method from the "ideal" line depends on the time the sample spends at the elevated temperature in zero field relative to the time it is influenced by an external field. This is supported by comparing the two sets of data of Figs. 1 and 2 for the two procedures of the Thellier experiment.

Because in the present experiments both the NRM and the subsequent PTRMs are produced by the same laboratory field, only time imbalance is possible between the spontaneous decay and the PTRM treatments at a particular temperature step. In an actual paleointensity determination there exists an additional possibility for non-symmetry. If the field producing the NRM differs from that producing the PTRMs, then different regions of the Néel diagram (NÉEL, 1949)—regions with different relaxation times—would be affected during the spontaneous decay steps than during the PTRM steps, leading to non-linear

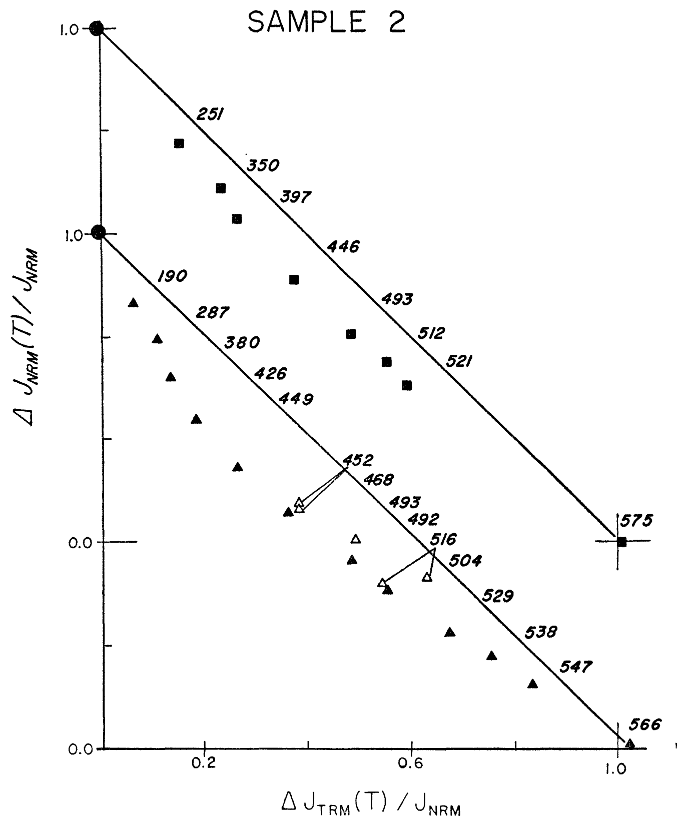

Fig. 3. PNRM-PTRM plots for sample 2. Figure format is identical to that of Fig. 1. Note that both data sets sag below the "ideal" line, although the curvature of the data of the "modified Coe" method is substantially greater.
$J_{TRM} = 0.14 \times 10^{-3}$ emu/g for $h_L = 0.46$ oe.

behaviour, even for the case where the time balance between the two treatments was maintained. The difference in the field intensities needed to produce observable non-linearities is yet to be determined. However, the above reasoning suggests that if the NRM field is much greater than the PTRM field, $|\boldsymbol{h}| \gg |\boldsymbol{h}_L|$, then the PNRM lost would be relatively greater than the PTRM that is measured after a particular temperature step, and the PNRM-PTRM curve would be concave up. That is, PNRM/PTRM$>|\boldsymbol{h}|/|\boldsymbol{h}_L|$, because a greater fraction of the remanence of the higher field NRM has relatively shorter relaxation times than the weaker field TRM (NÉEL, 1949). Thus the higher field NRM is relatively more susceptible to spontaneous decay than the smaller field PTRM to high-temperature VRM. Conversely, if the NRM field is much smaller than the PTRM field, $|\boldsymbol{h}| \ll |\boldsymbol{h}_L|$, then PNRM/PTRM$<|\boldsymbol{h}|/|\boldsymbol{h}_L|$ for a particular temperature step, leading to a PNRM-PTRM plot that is concave down. Therefore, to maximize the possibilities for linear PNRM-PTRM plots, the laboratory field used to impart PTRMs should be chosen to be as close as possible in intensity to the NRM-producing field.

Figure 3 shows that sometimes even the Coe method is not sufficient to produce linear PNRM-PTRM plots for samples containing a significant fraction of multidomain particles. Both sets of data are concave up with respect to the "ideal" line. For the Coe method a linear approximation for the data through the 493°C point has a slope of -1.22, whereas the ratio of initial to final points is -0.99 ± 0.01. The uncertainty represents the deviation from the mean of two determinations. The curvature associated with the data obtained by the "modified Coe" method is substantially greater than that of the Coe version. A linear least squares fit for the data through the 452°C point (solid triangles) has a slope of -1.50, whereas the ratio of initial to final points is -0.97. The non-linear behaviour for the Coe method is attributed to the presence of multidomain particles (LEVI, 1975), and the greater curvature of the "modified Coe" method is explained by the enhanced spontaneous decay.

For brevity, data of only three samples are presented above, although a total of ten samples were studied, Five of these additional samples were composed entirely of submicron grains and their PNRM-PTRM plots, obtained by the Coe method of the Thellier experiment, are as linear as the plots for samples 8 and 7. The PNRM-PTRM plots of the "modified Coe" method for these samples sag below the "ideal" line, with one possible exception. One sample containing rod-shaped (axial ratio 8 : 1) single domain magnetite particles was only incompletely studied by the "modified Coe" method: Three points (through 516°C) covering only above one third of the remanence are essentially linear and very close to the "ideal" line. The remaining samples contain varying fractions of grains larger than 1 μm. For these samples the PNRM-PTRM plots of the Coe method sag below the "ideal" line — the curvature increases with an increasing fraction of large particles. The PNRM-PTRM plots of the "modified Coe" method for these samples also sag below the "ideal" line, but their curvature is always substantially greater than the data obtained by the Coe method.

5. Conclusions

Of the two methods described here for the Thellier experiment, only the Coe method produces linear PNRM-PTRM plots for single domain and pseudo-single domain particles, because it maintains the symmetry between high-temperature VRM and spontaneous decay, while for the "modified Coe" method the spontaneous decay is enhanced. For multidomain remanence, even the Coe method yields a non-linear, concave up, PNRM-PTRM plot.

To avoid possible misunderstanding of the data of Figs. 1–3, it is advocated that the ratio of the end points, NRM/TRM, be *not* used for obtaining reliable paleointensities. In the above experiments both the NRM and TRM fields are known, and the samples and their magnetic properties are well characterized. This is in sharp contrast with actual paleointensity studies where the NRM field is sought and where the magnetic mineralogy and the chemical stability fields during the heatings are not usually known. In addition, Coe and Grommé (1973) have convincingly shown that a major source for non-ideal behaviour in paleointensity studies is caused by alterations in the samples' TRM properties upon heating in the laboratory. Paleointensity determinations utilizing only the ratio of NRM/TRM provide no means of evaluating the reliability of the results. Self-consistency checks, such as are provided by the Thellier experiment, are imperative for reliable paleointensity determinations.

I am grateful to Professor Ronald T. Merrill, my thesis advisor, for his encouragements and many valuable discussions, while this research was being done. Professor Minoru Ozima suggested the preparation of this manuscript. Professors Subir Banerjee, Minoru Ozima, and Mituko Ozima critically read the manuscript and their comments and suggestions have substantially improved it. Funding was provided by the National Science Foundation (NSF GA-27570 and DES 72-01541-A01) and by NASA (NGR-24-005-248).

REFERENCES

Coe, R.S., Paleointensities of the earth's magnetic field determined from Tertiary and Quaternary rocks, *J. Geophys. Res.*, **72**, 3247–3262, 1967 a.

Coe, R.S., The determination of paleointensities of the earth's magnetic field with special emphasis on mechanisms which could cause nonideal behavior in Thelliers' method, *J. Geomag. Geoelctr.*, **19**, 157–179, 1967 b.

Coe, R.S. and C.S. Grommé, A comparison of three methods of determining geomagnetic paleointensities, *J. Geomag. Geoelectr.*, **24**, 415–435, 1973.

LEVI, S., The effect of magnetite particle size on paleointensity determinations, *Phys. Earth Planet. Interiors*, 1975 (submitted).

NÉEL, L., Théorie du traînage magnétique des ferromagnétiques en grains fins avec applications aux terres cuites, *Ann. Géophys.*, **5**, 99–136, 1949.

THELLIER, E., Sur l'aimantation des terres cuites et ses applications géophysiques, *Ann. Inst. Phys. Glob. Paris*, **16**, 157–302, 1938.

THELLIER, E., Sur la thermorémanence et la théorie du métamagnétisme, *C.R. Acad. Sci., Paris*, **223**, 319–321, 1946.

THELLIER, E. and O. THELLIER, Sur l'intensité du champ magnétique terrestre dans le passé historique et géologique, *Ann. Geophys.*, **15**, 285–376, 1959.