Rock magnetic properties of an An-ei Lava, in Sakurajima Volcano
—Application to experimental study of geomagnetic paleointensity—

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Abstract

Magnetic mineral of An-ei lava in Sakurajima volcano that shows self-reversal pTRM (partial thermo-remanent magnetization) in narrow range of temperature was determined by X-ray diffraction method. It is cleared that magnetic mineral responsible to NRM (natural remanent magnetization) is not hemoilmenite but titano-magnetite Fe₃₋ₓTiₓO₄ of which molecular fraction of ulvo-spinel x=0.25~0.30. A new version of the Thelliers’ method of geomagnetic paleointensity (Zheng’s version) was applied in which pTRM was directly measured. Result obtained from the standard Thelliers’ method of the Coe’s version of the Thelliers’method was also shown. Experiments on thermal analysis of saturation magnetization and hysteresis were conducted from room temperature to liquid N₂ temperature, which suggested that the titano-magnetite of single domain was the main magnetic carrier.

Key words: An-ei lava, XRD, geomagnetic paleointensity, Coe’s version of Thelliers’ method, Zheng’s version of Thelliers’ method.

1. Introduction

An-ei lava (sample No. SF17) erupted in 1779 is one of the Sakurajima volcanic rocks reported by Ueno et al. (2004) which presented a partial self-reversal magnetization that might be caused by interaction between the two phases of titano-magnetite at double ranges of 245℃~260℃ and 330℃~340℃ (Fig. 1).

X-ray diffraction method (XRD) was carried to identify the magnetic mineral. The result documented the absent of hemo-ilmenite, a well reported carrier of self-reversal magnetization.

Hysteresis parameter was determined by measurement of hysteresis loop between...
liquid $N_2$ and 100°C to see the temperature dependence of the effective domain size of main magnetic mineral.

Paleointensity of the geomagnetic field was determined by Thelliers' method of both versions of Coe's and Zheng's (Zheng et al. 2004) to detect the effect of self-reversal pTRM appeared in narrow range of temperature.

Sample is collected in Furusato-machi Kagoshima City, Kyusyu, Japan. The sampling site was located at 31°33'00"N, 130°39'45"E. The detailed characteristics of NRM of the same sample are shown in Ueno et al. (2004).

2. Magnetic properties of the sample at low temperature

Thermal analysis of saturation magnetization (J$_s$) and hysteresis were performed by using a Vibrating Sample Magnetometer (VSM) made by Riken Denshi Co. at the magnetic field of 1000 mT (Fig. 2). The J$_s$ curve shows characteristic point of temperature at $-120^\circ$C $\sim -150^\circ$C, almost the same temperature as the Verwey point of magnetite ($-143^\circ$C).

The hysteresis parameters are plotted in Day diagram (Fig. 3). Results from Izu-Oshima basalt (1986 eruption) and Haruna dacitic lava (Hutatudake eruption) are also shown in the diagram. In the lower temperature, single domain becomes more effective as carrier of magnetism.
3. Result of XRD analysis

XRD analysis was performed at Hiroshima University to identify the magnetic mineral. The calibration of the X-ray diffraction meter was done by Si powder. In contrast to clear presence of titano-magnetite peaks, the peak of the ilmenite-
hematite series could not be detected. At the same time, it is cleared that magnetic
mineral responsible to NRM was a titano-magnetite Fe_{(6-x)}TiO_4, in which molecu-
lar ratio x of ulvo-spinel series was calculated to be x=0.25−0.30 based on the
linear assumption of x with lattice distance. Small x agrees with the result of
Js-T analysis that the characteristic temperature is close to the Verwey point of
magnetite. XRD data is shown in Fig. 4.
4. Paleointensity by Coe's version of Thelliers' method

Coe's version (Coe, 1967) of the standard Thelliers’s method was carried out in the nitrogen atmosphere with pTRM test. Sample was cut into a disk shape of 1 inch in diameter and 1 cm in thickness. It took about 1 hour in one cycle of heating and cooling including 10 minutes of hold time at the highest temperature level. Heating and cooling were carried in zero magnetic field at the temperature so called demagnetization step, then magnetized in the laboratory field parallel to NRM direction under the same temperature level to room temperature. These steps were carried out from room temperature to the Curie temperature with interval of 20°C~50°C. Paleointensity estimated by Williamson’s method (Kono and Tanaka, 1984) was 46.0 μT with data between room temperature and 204°C as shown in Arai diagram (Fig. 5). If data between room temperature and 240°C (self-reversal temperature) was used, the intensity was calculated to be 41 μT.

5. Paleointensity by Zheng’s version of Thelliers method

To reject data from the temperature block that shows inconsistency between NRM and TRM, Zheng’s version of Thelliers method was applied.

In Zheng's version, after thermal demagnetized at the temperature level Tc, pTRM (Tc, Tc−1) was acquired by heating from room temperature T0 up to Tc, cooling in artificial field perpendicular to the NRM direction to Tc−1 and without field back to T0. In the present case, the artificial field applied was 50 μT. Measurement of pTRM and NRM was carried after the 10 mT alternative current field demagnetization (AFD) to reduce the contribution of lower coercivity grains. Effectuality of
Fig. 6 Zheng version of Thelliers' method

Fig. 6-(1) Difference between before and after 10 mT AC demagnetization
- Blue……pTRM before AF demag.
- Red……pTRM after AF demag.

Fig. 6-(2) Apparent and corrected paleointensity of NRM
- Black open circle ...... δNRM / δT
- Red line ...... pTRM / δT
- Blue solid circle ...... Apparent paleointensity
- Red triangle ...... Corrected paleointensity

Fig. 6-(3) Apparent and corrected paleointensity of artificial TRM
- Black solid circle ...... δTRM / δT
- Green line ...... pTRM / δT
- Green solid circle ...... Apparent paleointensity
- Big green solid circle ...... Plateau paleointensity used for correction

Fig. 6-(4) Difference of pTRM between NRM and artificial TRM
- Red……pTRM of NRM
- Green……pTRM of artificial TRM
AF demagnetization was shown in Fig.6-(1) in which pTRM before with after demagnetization was compared. Paleointensity calculated in every temperature interval \((T_i, T_{i-1})\) were shown with blue solid circle in Fig.6-(2).

The same specimen was used for TRM test as shown in Fig.6-(3). TRM was acquired by cooling from the maximum demagnetization temperature level \(T_m\) to room temperature \(T_s\) in an artificial field of 50 \(\mu\)T parallel to the direction of NRM. Artificial TRM was then thermal demagnetized and applied pTRM with the same procedure and temperature intervals as the case of NRM. Intensity of artificial TRM, that should be 50 \(\mu\)T, calculated in every temperature interval \((T_{i-1}, T_i)\) were shown with solid green circle in Fig.6-(3).

If NRM was a thermal remanence originated, the attendance of NRM loss curve and that of TRM should be coincident to each other. And also if the thermal mineralogical alteration in experiment was not occurred, the spectrum of pTRM from NRM (1st run) and those from artificial TRM (2nd run) should be coincident to each other, because both were applied in the same field of 50 \(\mu\)T. In Fig. 6-(4), the spectrum of pTRM in both experiments was compared. In this case, both the pTRM spectra and NRM and TRM losses from 140°C to 200°C could be regarded as coincident to each other. Between 140°C and 200°C, NRM was seem pure thermal magnetization originated. Large difference between NRM and artificial TRM produced under 140°C, was caused by the large error of measurement because of the small pTRM at low temperature as well as by the chemical change in thermal experiment.

Plateau of the calculated artificial TRM intensity in 140°C~200°C noted with large green circle in Fig.6-(3) was divided by 50 \(\mu\)T to get correction rate. Corrected paleomagnetic field, shown with red triangle in Fig.6-(2), can be obtained from divided plateau noted with large blue solid circle divided by the correction rate. The average of the corrected paleomagnetic field is 44.3±0.6 \(\mu\)T.

The detailed procedure of the measurement by Zheng’ version of Thelliers’ method is illustrated in Fig. 7.

Fig.8 is the photograph of observation by electron microscope. Small magnetite of under 1 \(\mu\)m could be the particle of single domain or pseudo-single domain titano-magnetite.

In this study, paleointensity of SF17 was calculated under 200°C by both versions of Coe’s and Zheng’s, that is, under the double partial self-reversal ranges of 245°C~260°C and 330°C~340°C. SF17 was not a suitable sample to detect the effect of self-reversal pTRM appeared in narrow range of temperature on paleointensity study.
6. Results and Conclusion

Magnetic characters on An-ei lava in Sakurajima volcano that shows self-reversal pTRM in narrow range of temperature were studied. Hemo-ilmenite could not be found in XRD analysis. Main magnetic mineral was titanomagnetite $\text{Fe}_{3-x}\text{Ti}_x\text{O}_4$ of mol ratio of ulvo-spinel $x$ to be 0.25~0.30. The grain sizes are dominated by pseudo-single domain. Zheng's version of Thelliers' method gave reasonable palaeointensity of $44.3\pm0.6\mu\text{T}$. In Zheng's version, pTRM was directly acquired. The effect of self-reversal pTRM could not be found in palaeointensity study of SF17,
because the best paleointensity of SF17 was calculated under 200°C, the temperature under the double partial self-reversal ranges, in both methods of Coe's and Zheng’s.

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References
要 旨

上野直子・鄭 重・佐藤高晴：桜島安永溶岩の岩石磁気一古地球磁場強度実験への応用一

外部磁場を獲得する温度（ブロッキング温度）が異なるチタノマグネタイト相が近接して存在すると、狭い温度範囲で自己反転残留磁化が獲得されることがUeno他（2004）で確認された。その際に用いた桜島安永溶岩（試料番号SF17）について、基礎データとして液体窒素温度までの低温度Js-T、低温時のヒステリシス、XRD分析を行い、さらに古地球磁場強度測定に及ぼす影響を考慮した。古地球磁場強度測定では、通常使われているテリエ法Coe Versionの他に、外部磁場に比例しない残留磁化を獲得した温度範囲の残留磁化を排除して、部分熱残留磁化と自然残留磁化が比例する温度区間だけを使って古地球磁場強度を求める新しい方法（鄭Version）を用いた。

液体窒素までの低温中で振動試料型磁力計を用いて10kGの磁場中における飽和磁化の温度変化と約50℃おきのヒステリシスを測定した。ヒステリシスパラメーターをもちいたDay Plotは単磁区を含む擬似単磁区のチタノマグネタイトが主な磁性鉱物であることを示している。

磁石で抽出した強磁性鉱物を用いたX線回折（X-Ray Diffraction）ではイルメナイト−ヘマタイト系列の信号がないことを確認し、狭い温度範囲での自己反転残留磁化は棒名のヘモイルメナイトによる自己反転とは異なる反転獲得機構であることを確信した。ウルボスピネル−マグネタイト系列のチタノマグネタイトの回析線が検出された。このチタノマグネタイトの回析線はウルボスピネルよりもマグネタイトの回析線の方が近い。格子距離がTiの分子比と直線的に比例すると仮定すると、Tiの分子比はx=0.25～0.30であった。

古地球磁場強度測定に使用されるテリエ法はNRMがTRM起源であり、古地球磁場強度に比例していることが前提である。常温からキュリー温度まで、20度から50度の温度間隔で、無磁場中の加熱後の残留磁化と同一温度での一定磁場での加熱後の残留磁化を用いてアライダイアグラムから計算した場合（テリエ法Coe Version）は常温から約200度までのデータから46μTが得られた。

新しく開発した、鄭Versionテリエ法では、まずNRMを熱消磁した温度区間にその区間の50μTの定磁場で部分熱残留磁化（pTRM）をNRMに垂直になるように直接つけ（Coe Versionでは室温から熱消磁温度までつける）、この温度区間に消磁されたNRMと獲得されたpTRMから温度区間ごとに見かけの古地球磁場強度を計算する。次に、50μTの定磁場でNRM方向に室温からNRMの最高の熱消磁温度まで人工的TRMをつけて、この人工的TRMをNRMの場合と同様に温度区分熱消磁および定磁場での部分熱残留磁化の直接付加から人工的TRMの古地球磁場強度を計算する。この人工的TRMの古地球磁場強度は50μTのはずなので、計算値と50μTの比率の値を補正項としてNRMによって得られた見かけの古地球磁場強度を補正する。NRMおよび人工的TRMの区間別熱消磁後につけた部分熱残留磁化同士を比較して差が大きく出る温度区間の値は実験中に化学変化が起きたためと考え除去する。有意差が出ない温度以下で新しい試料で再度実験すればより説得力のある結果が得られる。この鄭方法で求めた古地球磁場強度は44.3±0.6μTであった。これは観測値から期待される44μTと一致する。SF17では、
自己反転温度区間が古地球磁場強度測定実験範囲温度よりも高いためCoe方法と鄭方法の差がでなかった。鄭方法を用いると、どの温度範囲でNRMがTRM起源なのか、すなわち火成岩をもちいた古地球磁場強度測定実験におけるNRMの有効温度範囲を決定することが出来る。この新法の難点は手順が増えること、温度制御が正確な炉が必要なことの他に、NRMと比較して少量のpTRM（特に低温部では少ない）から有意義な結果を出すためには試料の設置方向の再現性など誤差の減少に細心の注意が必要である。しかし、火成岩起源の試料から正しい値を得るためには、この新法で古地球磁場強度を求めるべきである。

新しい古地球磁場強度測定法の具体的な手順はFig.6-(1), Fig.6-(2), Fig.6-(3), Fig.6-(4), に示した。
1. NRMを室温からTₘまで熱消磁して、残留NRMを10mTで交流消磁した後で測る。
2. 室温からTₘまで無磁場で加熱して、冷却温度区間Tₘ-1〜T₂のみに50μTの定磁場でNRMに垂直になるように部分熱残留磁化を直接付加する。温度Tₘ-1から室温までは無磁場で冷却する。10mTで交流消磁して残留pTRMを測る。Fig.6-(1)は交流消磁前後の比較。交流消磁前を青、後を赤棒グラフで示した。
3. 温度区間ごとに中空黒丸と赤棒グラフ値から見かけの古地球磁場強度を計算する。見かけの古地球磁場強度はFig.6-(2)に中塗青丸で示した。
4. 室温から2.の実験の最高温度までNRM方向に人工的なTRMを付加して、NRMの一部を人工的なTRMで置き換える。
5. TRMをNRMに見立てて、上記1.〜3.までの実験をする。
   Fig.6-(3)の中塗緑丸は人工的TRMの見かけの古地球磁場強度で50μTになるはずの値である。
6. NRMに対するpTRMと人工的TRMに対するpTRMは実験中に化学変化が起きずにNRMがもとのTRM起源のみなら同じ値になるはずなので、比較する。Fig.6-(4)で赤と緑が一致するとき試料に化学変化がなかったと言える。また、NRMの強度変動微分曲線がTRMのと同じ傾向ならNRMがTRM起源であるといえる。差が大に出る温度区間のデータは使えない。低温区間での差はTRMやNRMの強度差が大きいことに、高温区間でTi移動などの化学変化が生じたことが考えられる。
7. Fig.6-(4)で赤と緑が一致する温度区間についてFig.6-(3)の中塗大緑丸の平均値を算出する（Plateau F）。Plateau Fと50μTの比の値を補正項とする。
8. 補正項を用いて7.の温度区間について3.の見かけの古地球磁場強度（中塗大青丸）を補正する。補正後の古地球磁場強度をFig.6-(2)に赤三角で表示した。この平均値が新法による古地球磁場強度である。