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Identification of magnetic minerals carrying NRM in a pyroclastic-flow deposit: implication in the lowtemperature magnetic component

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1. Introduction

Many paleomagnetic and rock-magnetic researches have been carried out on volcanology. In particular, the method on the basis of the theory of thermoremanent magnetization (TRM) has made a significant contribution to determine the origins of volcaniclastic deposits. Aramaki and Akimoto (1957) first distinguished pyroclastic-flow deposits from debris-flow deposits by measuring directions of initial natural remanent magnetization (NRM). Subsequently, Hoblitt and Kellogg (1979) established the method adopted progressive thermal demagnetization (PThD). This method made possible quantitative estimates of emplacement temperatures of hot volcaniclastic deposits, especially pyroclasticflow deposits. Therefore, it has been used in many researches (e.g. Kent et al. 1981; Zlotnicki et al. 1984; McClelland and Druitt 1989; Tamura et al. 1991; Clement et al. 1993; Bardot 2000; Saito et al. 2000).

One problem underlying the method, however, has often troubled us, as Hoblitt and Kellogg (1979) already pointed out. In this method, emplacement temperature estimation can be made by clarifying the unblocking temperature of NRM that was acquired during and after the emplacement of a pyroclastic-flow. This is based on the assumption that NRM originates from TRM. If the direction of Earth's magnetic field during cooling of the pyroclastic-flow deposit differs from that of the present field, TRM is probably the origin of remanence that grouped about the field direction during cooling. However, if the field during cooling is parallel to the present field, the origin of NRM may not be TRM but viscous remanent magnetization (VRM). In the latter case, it is impossible in theory to estimate the emplacement temperature only by this method.

Geological setting

Saito et al. (2000) also confronted this problem in studying the Ikeshiro pyroclastic-flow deposit distributed on the northwestern foot of Yufu volcano, Kyushu Island, Japan. Although they have surmounted it by presenting another cognate volcaniclastic deposit without taking VRM and by comparing the direction of the low-temperature component with that of Earth's magnetic field, these are not enough to say that NRM originates not from VRM but from TRM. In this paper, we carried out paleomagnetic and rock-magnetic experiments on samples from the Ikeshiro pyroclastic-flow deposit, to examine the origin of NRM in terms of magnetic minerals. First, we made paleomagnetic experiments, including PThD and progressive alternating field demagnetization (PAFD). Based upon this result, we classified our samples into two types and carried out three kinds of rock-magnetic experiments on samples of each type to identify magnetic minerals and their grain size in a rock. It is because magnetic minerals and their grain sizes are important factor in the acquisition of VRM.

2. Geological setting

Yufu volcano is one of the most active Quaternary stratovolcanoes in the Hohi volcanic zone in central Kyushu Island (Kobayashi 1984; Hoshizumi et al. 1988). It is blanketed with one summit lava and eight lateral volcano lavas, one of which located on the northern slope is called the Ikeshiro lava dome. In addition, a debris-avalanche deposit, pyroclastic-flow deposits, debris-flow deposits and fan deposit were spread at the base of the volcano. The activity of Yufu volcano started from more than 35,000 years ago (Kobayashi 1984). The last activity brought the Ikeshiro lava dome and the summit lava about 2,000 years ago. They collapsed repeatedly and generated many block-and-ash flows. One of them emplaced on the northwestern foot is called the Ikeshiro pyroclastic-flow deposit (Fig. 1). It is dated as 2090 ± 90 years B.P. by ¹⁴C method using charcoal (Okuno et al. 1999).

The Ikeshiro pyroclastic-flow deposit is a non-welded block-and-ash flow deposit. It is composed of hornblende andesite dense juvenile clasts and matrix of the same lithology. Some of the clasts include vesicular autolith with diktytaxitic structure. The Ikeshiro pyroclastic-flow deposit is more than 10 m thick and consists of at least 3 flow units. Each unit is separated by fine ash layer. The bottom of top unit is finesdepleted as well as rich in clasts; it is similar to lithofacies of a ground layer (Walker et al. 1981). The top of central unit contains pipes filled in lithic fragments (e.g. Francis 1993). The clasts are mostly poorly vesiculated and commonly oxidized in various degrees. Unoxidized clasts are mostly grayish or light grayish color similar to fresh lava. On the other hand, oxidized clasts usually have reticular cracks with reddish tints. The sizes of the clasts range from a few centimeters to over 10 meters.



Fig. 1

Simplified geologic map of the north side of Yufu volcano after Saito et al. (2000). The location of samples is also shown. Triangles are Quaternary volcanoes in Kyushu. A, Aso; K, Kuju; S, Sakurajima; T, Tsurum; U, Unzen.

3. Paleomagnetic experiments

3.1 Sampling and experimental procedure

Paleomagnetic samples were collected at one outcrop from the central unit of the Ikeshiro pyroclastic-flow deposit. Location of the sampling site is also shown in Fig. 1. Fourteen lava clasts were oriented using a magnetic compass. They are at most 15 cm in maximum diameter. In the laboratory, 25 mm diameter and 22 mm height specimens were cut from the oriented clasts. NRM of each specimen was measured using a spinner magnetometer (Natsuhara-Giken SMM-85). Specimens were demagnetized thermally and magnetically in a magnetic field is less than 1 μ T around the measurement instruments. PThD was carried out up to 590°C or 650°C in the air using an electric furnace (Natsuhara-

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Giken) in a four layer mu-metal magnetic shield. Residual field is less than 5 nT. Low-field magnetic susceptibility of specimens was measured using a Bartington MS2 magnetic susceptibility meter at each thermal demagnetization step. PAFD was carried out up to a peak alternating field of 100 mT or 150 mT using a three-axis tumbler system (2 G). Demagnetization results were plotted on vector end point diagrams (Zijderveld 1967). On the diagrams, we determined the components of remanent magnetization and applied principal component analysis to each component (Kirschvink 1980). A stable magnetic component is defined as follows, (1) linear segment consists of more than three vector end points and (2) maximum angular deviation (MAD) is less than 15°.

3.2 Classification by paleomagnetic results

As a result of PThD and PAFD, all specimens have two or more stable magnetic components. In PThD, the components removed at the lowest demagnetization level show in better grouping of the direction (Dec = -0.8° , Inc = 32.1° , $\alpha 95 = 10.1^{\circ}$, Fig. 2; Saito et al. 2000). The maximum unblocking temperature of this component ranges from 200°C to 450°C. This direction is close to the direction of the present Earth's magnetic field at this location (Dec = -6.5° , Inc = 47°). Initial NRMs and the other magnetic components are scattered.

Fig. 2

Equal-area projection showing directions of the low-temperature magnetic component (\bullet), their mean direction (\blacksquare) and associated 95 per cent confidence circle for the Ikeshiro pyroclastic-flow deposit. It also shows the direction of the present Earth's magnetic field (\blacklozenge) and that of 2,000 years ago (\blacklozenge) after Ohno et al. (1991).



When we compared the trajectories on the diagrams from PAFD with those from PThD, the specimens were classified into the following two types. We name the former Type A, the latter Type B. Seven samples were Type A and six were Type B.

Type A

The trajectories of vector end points on the diagrams of PThD are similar to those of PAFD (Fig. 3a). The low-temperature magnetic component is removed at low alternating field (5-10 mT). For example, IK24 has three magnetic components. First component is demagnetized below 250°C and 5 mT. Second is demagnetized below 425°C and 12.5 mT. The last is demagnetized above 450°C and 17.5 mT. This indicates that magnetic carriers of the low-temperature and high-temperature



Fig. 3

Vector end point diagrams of PThD and PAFD results for typical samples of the Ikeshiro pyroclastic-flow deposit. IK24 (a) is example of Type A, while IK22 (b) and IK4 (c) are examples of Type B. One specimen of IK22 was thermally demagnetized up to 680°C after PAFD of 150 mT (b-2). Solid and open circles are projections onto the horizontal and N-S vertical planes, respectively.

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component possess low and high coercivity, respectively. Complete demagnetization by 580°C and 100 mT indicates that the main magnetic carriers of the remanence are magnetite. There is no indication of hematite

Type B

The trajectories on the diagrams of PThD differ from those of PAFD (Fig. 3b, c). The low-temperature components could not be removed up to 150 mT. For example, IK22 has three components. In PThD, first component (Dec = 9.5° , Inc = 23.3° , MAD = 3.3°) is demagnetized below 325° C, second (Dec = 202.1° , Inc = -28.1° , MAD = 1.4°) is between $325^{\circ}C$ and $520^{\circ}C$, and the last (Dec = 213.0° , Inc = -23.1° , $MAD = 0.9^{\circ}$) is above 520°C (Fig. 3b-1). On the contrary, only one magnetic component (Dec = 200.3° , Inc = -26.6° , MAD = 0.7°) is removed in PAFD of 150 mT (Fig. 3b-2). The direction of this component is closest to that of the medium-temperature component. Then, we carried out PThD to this specimen up to 680°C after PAFD. As a result, we could remove the low-temperature component ($Dec = 3.7^{\circ}$, Inc = 23.2° , MAD = 1.9°) below 325° C and the high-temperature component (Dec = 210.4° , Inc = -19.9° , MAD = 0.5°) above 550° C. Between 325°C and 550°C, the medium-temperature component (Dec = 200.3°, Inc = -28.5° , MAD = 2.5°) is demagnetized again. This indicates that magnetic carriers of the low-temperature and high-temperature component possess high coercivity, while the medium-temperature component is mostly carried by magnetic mineral with low coercivity and partly carried by that with high coercivity. Another example is shown in Fig. 3c. IK4 has three magnetic components in PThD but none of the components can be removed in PAFD. This indicates that the coercivity spectra of each component overlap almost completely. IK4 is holocrystalline-porphiritic autolith probably due to cognate origin. Therefore, it is likely that the properties of magnetic minerals in IK4 differ from other samples. This may be the cause of the overlapping.

3.3 Magnetic minerals presumed from paleomagnetic results

On the basis of paleomagnetic results, we presumed magnetic minerals carrying NRM of each type as follows.

Type A

The low-temperature component is probably carried by multidomain (MD) grains of magnetite because of its low coercivity. On the other hand, the high-temperature component is perhaps carried by single-domain (SD) grains of magnetite. Paleomagnetic results do not indicate any other minerals carrying the remanence.

Type B

The low-temperature component is carried by magnetic mineral with

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high coercivity (above 150 mT). The medium-temperature component is mostly carried by magnetite and partly carried by magnetic mineral with high coercivity. The high-temperature component is carried by hematite. It is possible for magnetic mineral carrying the low-temperature component to be SD grains of titanomagnetite, titanohematite, goethite or pyrrhotite. From these results only, we cannot determine which is the main carrier of the remanence.

4. Rock-magnetic experiments

Three kinds of rock-magnetic experiments were conducted in a nonshielded room at Kyoto University. They are (1) progressive acquisition of isothermal remanent magnetization (IRM), (2) the modified Lowrie-Fuller test (Johnson et al. 1975) and (3) PThD of a three-component IRM (Lowrie 1990). By the first and second experiment, we can know the kinds and grain sizes of magnetic minerals included in the rocks. By the last experiment, we can find the unblocking temperatures of each IRM component with different coercivity. We selected IK24 (Type A) and IK22 (Type B) as rock-magnetic samples from fourteen clasts because these two samples are typical of each type. Remanent magnetization of each specimen was measured using a spinner magnetometer (Natsuhara-Giken SMM-85).

4.1 IRM acquisition

IRM was given with an electromagnet (Eiko Electric Ltd.) up to maximum fields of 1.7 T. The normalized IRM acquisition curves for both samples (Fig. 4) rise very steeply initially and reach over 95 % by 0.1 T. This indicates that magnetite is the predominant magnetic mineral in these samples. While IK24 achieve complete saturation by 0.4 T, the curve of IK22 continues to climb gently and does not saturate by 1.7 T. This indicates that IK22 includes magnetic mineral with high coercivity besides magnetite.



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4.2 The modified Lowrie-Fuller test

Anhysteretic remanent magnetization (ARM) was produced by applying a direct field of 0.1 mT in a peak alternating field of 100 mT. Saturation isothermal remanence (SIRM) was produced in a direct field of 1.7 T. PAFD was carried out up to a peak alternating field of 100 mT using a three-axis tumbler system (Natsuhara-Giken). ARM and SIRM of IK22 could not be completely demagnetized by 100 mT (Fig. 5b). This indicates that IK22 includes magnetic mineral with high coercivity. Therefore, we cannot discriminate the dominant grain size of magnetite. On the other hand, IK24 shows MD-type results (Fig. 5a). The normalized PAFD curve of ARM always lies below that of SIRM. Exponential shape of the curve of ARM also indicates the predominance of MD grains. However, the demagnetization curve of SIRM is not a typical MD-type. There is a small initial superexponential plateau extending to 10 mT as is seen in the curve of SD-type, while the curve has the exponential form above 20 mT. This suggests that some grains act like SD grain as to SIRM while they act like MD grain as to ARM. It is possible that pseudo-single-domain (PSD) grain act like this (Dunlop 1983), although precise property of PSD grain is still far from certain.



4.3 The three-component IRM

The three-component IRM was produced by applying a direct field of 1.7 T along the *z* axis, followed by 0.4 T along the *y* axis and finally 0.12 T along the *x* axis. Then, PThD was carried out up to 600°C for IK24 and 680°C for IK22 in the air using an electric furnace (Natsuhara-

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Giken) in a three-layer mu-metal magnetic shield. Residual field is less than 7 nT. Low-field magnetic susceptibility of each specimen was also measured using a Bartington MS2 magnetic susceptibility meter at each thermal demagnetization step. Demagnetization results are shown in Fig. 6 by plotting and evaluating the thermal decay of each component separately. Most of the remanence of IK24 is soft (<0.12 T) coercivity fraction and little magnetization remains with hard (0.4-1.7 T) fraction (Fig. 6a). The soft and medium (0.12-0.4 T) coercivity fraction demagnetizes mostly between 400°C and 560°C and completely by 580°C. Little demagnetization occurs out of this range. This is strong evidence that magnetite is the predominant magnetic mineral. In particular, MD grains are predominant because the soft fraction is much more abundant than the medium fraction. There is no indication of hematite and any other magnetic minerals. On the other hand, each fraction of IK22 shows distinct unblocking temperatures (Fig. 6b). The soft fraction demagnetizes mostly between 400°C and 580°C. This indicates that main magnetic carrier of the soft fraction is MD magnetite. On the contrary, the medium and hard fraction resist above 580°C and completely demagnetizes by 680°C. This indicates that part of the medium and hard fraction are carried by hematite. The unblocking temperature of the medium fraction has broad range mainly from 320°C to 620°C. This is thought to be because this fraction is carried by SD magnetite and hematite with low coercivity, therefore unblocking temperature of both minerals overlap. The hard fraction shows most remarkable result. The demagnetization curve abruptly drops between 160°C and 240°C, while the curve decays continuously above 240°C up to 680°C. This dropping does not occur in the other fractions. This indicates that the hard fraction is carried by not only hematite but also other magnetic mineral with high coercivity and low unblocking temperature.



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5. Discussion

As a result of paleomagnetic and rock-magnetic experiments, we found that Ikeshiro samples are classified into two types and each type includes different composition of magnetic minerals. In this chapter, we will discuss origin of the low-temperature magnetic component of the Ikeshiro pyroclastic-flow deposit in terms of magnetic minerals. In addition, we found that the classification corresponds to oxidation state of samples. At the last section, we will discuss volcanological implications of our results in terms of oxidization.

5.1 Identification of magnetic minerals

Three kinds of rock-magnetic experiments reveal properties of magnetic minerals included in the samples of each type. These properties are consistent with paleomagnetic results.

Type A

Magnetic minerals are mainly MD grains of magnetite and partly SD magnetite. Result of the modified Lowrie-Fuller test suggests existence of PSD magnetite. Hematite and other magnetic minerals are not included. This is as expected from paleomagnetic results.

Type B

Type B includes three kinds of magnetic mineral. Magnetite is predominant mineral according to the progressive acquisition of IRM. As for grain size, Type B is thought to include more SD grains than Type A does although we could not determine directly by rock-magnetic experiments. It is because NRM of Type B resists PAFD below 7.5 mT while Type A demagnetizes to a great extent (Fig. 3a-2, 3b-2). Hematite is also included. In addition to these two minerals, paleomagnetic results suggested existence of another mineral with low unblocking temperature and high coercivity. The three-component IRM reveals that its coercivity is more than 400 mT and it unblocks between 160°C and 240°C. Judging from this, the mineral is thought to be not SD titanomagnetite. goethite or pyrrhotite but titanohematite with $y \approx 0.45$. The coercivity is too high for SD titanomagnetite whose microscopic coercivity is less than 300 mT. Goethite cannot carry the remanence between 160°C and 240°C because the Curie temperature is 110-130°C. If this mineral is pyrrhotite, initial susceptibility must show some change due to breakdown of pyrrhotite into magnetite or hematite above 500°C. However, susceptibility change of IK22 is less than 10 % and there is no abrupt change. On the contrary, titanohematite for $0 \le y \le 0.5$ possesses high coercivity like hematite and the Curie temperature of $y \approx 0.45$ is about 240°C. Therefore, we conclude that Type B includes magnetite, hematite and titanohematite with $y \approx 0.45$.

5.2 Origin of the low-temperature component

On the basis of the magnetic minerals carrying the low-temperature magnetic component, we will discuss origin of the low-temperature magnetic component.

Type A

The low-temperature component is carried by MD grains of magnetite, while the other components are carried by SD grains or PSD grains of magnetite. MD grain of magnetite can generally acquire more VRM than SD grain does because of domain wall motion due to thermal activation (e.g. Dunlop and Özdemir 1997). MD grain possesses microscopic coercivity dominantly less than 20 mT, while SD and PSD grain possess higher coercivity. Microscopic coercivity of a grain is the lower, the larger viscosity coefficient becomes and the more VRM is acquired. The low-temperature component of Type A possesses coercivity less than 5-10 mT. Therefore it is quite possible that the low-temperature component of Type A originates from VRM.

Type B

The low-temperature magnetic component is carried by titanohematite with $y \approx 0.45$, while magnetite and hematite carry the remanence unblocked at higher temperature. We have little knowledge and experience on the VRM acquisition of titanohematite as far as the authors know. Now we assume that titanohematite can acquire VRM in the same way as hematite because titanohematite for $0 \le y \le 0.5$ is essentially antiferromagnetic with a weak parasitic ferromagnetism, like hematite. Hematite has a large range of stable SD grain-size, so a large portion of hematite is SD. Thus, we apply the SD theory to titanohematite. For assemblages of SD grains, the acquisition of VRM is essentially the inverse of magnetic relaxation. The relaxation time is directly related to the microscopic coercivity. High coercivity of titanohematite with $y \approx$ 0.45 brings long relaxation time. Long relaxation time prevents the grain from acquiring VRM. However, the relaxation time is also related to volume of the grain. If the volume is very small, the grain is affected by thermal activation and acquires VRM (Dunlop and Stirling 1977). This VRM is hard to PAFD because its microscopic coercivity is high. We do not know the grain size of titanohematite, so we cannot examine the possibility of hard VRM theoretically. However, according to PAFD of acquired VRM, part of VRM demagnetizes below 100 mT while part resists above 100 mT (Biguand and Prévot 1971). On the contrary the low-temperature component of Type B does not demagnetize by alternating field of 150 mT at all (Fig 3b-2). This suggests that the low-temperature component does not originate from simple VRM.

In addition, Saito et al. (2000) is the same opinion. They previously studied NRM of the Ikeshiro pyroclastic-flow deposit. They concluded that the low-temperature component originated not from VRM but from TRM for the following two reasons. (1) When they measured

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NRM of a debris-flow deposit cognate with the Ikeshiro pyroclasticflow deposit, there were not any magnetic components parallel to the present Earth's magnetic field. (2) Direction of the Earth's field during cooling was within 95 per cent confidence limit for the mean direction of the low-temperature component. The former is a good reason for TRM, although the latter reason is uncertain because the Earth's field about 2,000 years ago changes its direction about 10 degrees for a hundred year (Ohno et al. 1991).

In conclusion, the low-temperature component of Type B originates not from VRM but from TRM, while that of Type A possibly originates from VRM. If we try to examine the origin of NRM more closely, we need more experiment, for example the acquisition of VRM.

5.3 Volcanological implications

What do our results and conclusions mean volcanologically? When we go back to the basic, we can estimate the emplacement temperature. Maximum emplacement temperature of the Ikeshiro pyroclastic-flow deposit is thought to be about 450°C according to PThD. However, we think only this is uninspiring. Saito et al. (2000) ambitiously discussed activity of the Ikeshiro lava dome, which generated the Ikeshiro pyroclastic-flow, on the basis of paleomagnetic data and oxidation state of the clasts. Finally we will discuss generation of the Ikeshiro pyroclastic-flow deposit on the basis of our results and oxidation state of the clasts.

We classifies our samples under three degrees of oxidation (unoxidization, weak oxidization and strong oxidization) after Saito et al. (2000). Unoxidized samples consist of grayish groundmass and fresh phenocrysts. Strongly oxidized samples consist of reddish groundmass and weathered phenocrysts, especially broken down hornblende with opacite rim. Weakly oxidized samples are medium between the two and part of the groundmass is reddish. Five of seven Type A samples are unoxidized and the rest are weakly oxidized. Type B samples are strongly oxidized except for IK4.

As a result of our experiments, we found that unoxidized or weakly oxidized Type A includes only magnetite, whereas strongly oxidized Type B includes hematite and titanohematite besides magnetite. This suggests that oxidization caused each type to include different magnetic minerals because hematite and titanohematite are the higher oxidation state. We think this oxidization had been occurred at the Ikeshiro lava dome before generation of the Ikeshiro pyroclastic-flow. The clasts were sampled at random from the unit. If oxidization was occurred during or after the emplacement of the deposit, oxidization had to be occurred at random. This is, however, quite unreasonable and there is no evidence that such oxidization had been occurred. Oxidization had been already made before the emplacement of the deposit. In addition, existence of

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titanohematite with $y \approx 0.45$ indicates that the clast was quenched from very high temperature because titanohematite of intermediate composition is not an equilibrium phase at room temperature and it can be preserved only by rapid cooling from very high temperature (Lindsley 1976). This rapid cooling is happened during transportation of the pyroclastic-flow. Then, only lava dome is able to oxidize the clasts before generation of the pyroclastic-flow. Lava blocks exposed on the dome surface are strongly oxidized and hematite and titanohematite with $y \approx 0.45$ are produced by oxidization. On the other hand, lava blocks at the dome core are hardly oxidized. Collapse of the lava dome generates the Ikeshiro pyroclastic-flow and titanohematite is quenched and is preserved. This idea explains all results of this study consistently. We believe that a study of magnetic minerals will lead to more understanding of lava dome.

6. Conclusion

The following conclusions were reached as a result of this study.

1. Samples from the Ikeshiro pyroclastic-flow deposit are classified into two types. Type A demagnetizes magnetically in the same way as PThD, while Type B does not.

2. Samples of each type are carried by different magnetic minerals. Type A is carried by mainly MD magnetite and partly SD magnetite. Type B is carried by magnetite, hematite and titanohemtite with $y \approx 0.45$. The low-temperature magnetic component of Type A is carried by MD magnetite, while that of Type B is carried by titanohematite.

3. Although MD magnetite can acquire VRM easily, titanohemtite with $y \approx 0.45$ can hardly acquire VRM because of its high coercivity. Therefore, the low-temperature magnetic component of Type A possibly originates from VRM, while Type B originates not from VRM but from TRM.

4. On the basis of this result and oxidation state of samples, we think of generation of the Ikeshiro pyroclastic-flow deposit about 2,000 years ago as follows. (1) Initial magnetic minerals, perhaps magnetite, crystallize from magma. (2) The eruption begins and the Ikeshiro lava dome is formed. Lava blocks exposed on the dome surface (Type B) is strongly oxidized, while the blocks at the dome core (Type A) is hardly oxidized. At this stage, hematite and titanohematite with $y \approx 0.45$ is produced on the dome surface. (3) The dome collapses and generates the Ikeshiro pyroclastic-flow. Titanohematite is quenched and is preserved. (4) The Ikeshiro pyroclastic-flow is emplaced at about 450°C and titanohematite with $y \approx 0.45$ acquires TRM during cooling.

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