Magnetic Susceptibility of Ferromagnetic Minerals Contained in Igneous Rocks.*

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Abstract

The thermal change in susceptibility of ferromagnetic mineral grains separated from igneous rocks was measured by a ballistic method in a weak magnetic field. The thermal change of susceptibility was classified into four types. These types were discussed in connection with the condition under which the original rock was formed. The dependency of the Curie-point and the magnitude of the susceptibility upon the chemical constitution was examined, the content of TiO₂ in ferromagnetic mineral being especially taken into account. Besides, the relation between the magnitude of susceptibility and the grain size was obtained from the viewpoint of the magnetic property of small particles.

§ 1. Introduction

The magnetic properties of igneous rocks have been examined fairly in detail by a number of investigators (1), (2), (3), (4), (5), (6), especially in connection with the local geomagnetic anomalies and their changes. The magnetic properties of rocks, however, are due to those of ferromagnetic minerals contained in them, the other minerals scarcely contributing to the magnetism of rocks. In other words, the magnetism of rocks is composed of the ensemble of magnetization of small grains of rock-forming ferromagnetic minerals. So that the fundamental problem in rock-magnetism will be the general description of rock-forming ferromagnetic minerals from the physical standpoint in reference to their chemical and mineralogical characteristics.

In the present study, the change with the temperature in magnetic susceptibility of rock-forming ferromagnetic minerals, which were separated from various kinds of mother igneous rocks, was measured, and the results were analysed in relation to the chemical composition, grain size and mineralogical characteristics of these grains of minerals. The aims of the present study are chiefly the following two.

(1) The natural ferromagnetic minerals contained in igneous rocks are generally of chemical constitution different from a pure stoichiometric magnetite. It will be significant, therefore, to find the relations of the Curie-point and the magnitude of susceptibility to the chemical composition.

(2) On the other hand, it has been expected that the dimensions of grains play an important role upon the magnetic properties such as initial permeability and

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coercive force. (7) Since the grain sizes of those specimens which were dealt with in the present study range over some extent, the relation between the initial susceptibility and the grain size was also examined.

§ 2. Specimens

The specimens examined here are 15 in total and each of them is the magnetite grains, petrologically so-called for the sake of simplicity, separated by an electromagnetic separator from original igneous rocks which were collected from volcanic localities in Kwanto District of Japan. The mean diameters of 50 grains of each sample were measured by means of a microscope. The result shows that the grain sizes vary from about 30\(\mu\) to 400\(\mu\), the discrepancy depending on either they appear as phenocrysts or as microcrystals in groundmass in the original rocks. Their grain sizes, states of appearance in rocks and localities are shown in Table 1 together with the chemical constitutions which were analysed by I. Iwasaki and his colaborator. (3)

As will be assumed from the date of the chemical composition shown in

<table>
<thead>
<tr>
<th>No.</th>
<th>Locality</th>
<th>Petrological Description</th>
<th>Grain Size</th>
<th>(\text{Fe}_2\text{O}_3 + \text{FeO})</th>
<th>(\text{TiO}_2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Niizima</td>
<td>Phenocryst magnetite in biotite liparic</td>
<td>110(\mu)</td>
<td>63.01 31.46%</td>
<td>4.06 (4.06)%</td>
</tr>
<tr>
<td>2</td>
<td>Lava in Asigara</td>
<td>Phenocryst magnetite in hyp. horn. dacite</td>
<td>130</td>
<td>67.60</td>
<td>10.81(13.73)</td>
</tr>
<tr>
<td>3</td>
<td>Zybosi volc. Ito</td>
<td>Groundmass magnetite in olivine basalt</td>
<td>53</td>
<td>56.66</td>
<td>14.44(20.08)</td>
</tr>
<tr>
<td>4</td>
<td>Simohutagayama, Hakone</td>
<td>Phenocryst magnetite in hyp. aug. andesite</td>
<td>120</td>
<td>75.80</td>
<td>10.86(12.46)</td>
</tr>
<tr>
<td>5</td>
<td>Kaziya Yuga-wara</td>
<td>Phenocryst magnetite in hyp. dacite</td>
<td>140</td>
<td>74.44</td>
<td>10.24(12.08)</td>
</tr>
<tr>
<td>6</td>
<td>Yugawara</td>
<td>Phenocryst magnetite in aug. horn. dacite</td>
<td>130</td>
<td>69.84</td>
<td>8.73(11.05)</td>
</tr>
<tr>
<td>7</td>
<td>Karataki, Hakone</td>
<td>Phenocryst magnetite in aug. horn. dacite</td>
<td>110</td>
<td>75.71</td>
<td>12.17(13.79)</td>
</tr>
<tr>
<td>8</td>
<td>Hiroawara</td>
<td>Phenocryst magnetite in aug. bearing hyp. dacite</td>
<td>130</td>
<td>83.28</td>
<td>11.40(12.02)</td>
</tr>
<tr>
<td>9</td>
<td>Odawara</td>
<td>Phenocryst magnetite in pumice</td>
<td>280</td>
<td>88.13</td>
<td>6.43(6.79)</td>
</tr>
<tr>
<td>10</td>
<td>Taga volc.</td>
<td>Phenocryst magnetite in oliv. aug. andesite</td>
<td>200</td>
<td>75.60</td>
<td>10.73(12.34)</td>
</tr>
<tr>
<td>11</td>
<td>Haruna volc.</td>
<td>Phenocryst magnetite in hyp. horn. dacite pumice</td>
<td>390</td>
<td>53.15 31.93</td>
<td>6.82(7.33)</td>
</tr>
<tr>
<td>12</td>
<td>Sukumogawa Hakone</td>
<td>Groundmass magnetite in andesite</td>
<td>43</td>
<td>65.05</td>
<td>16.59(20.18)</td>
</tr>
<tr>
<td>13</td>
<td>Okata Oosima</td>
<td>Groundmass magnetite in tholeite</td>
<td>63</td>
<td>47.19 33.71</td>
<td>17.20(17.20)</td>
</tr>
<tr>
<td>14</td>
<td>Taga volc.</td>
<td>Groundmass magnetite in olivine basalt</td>
<td>46</td>
<td>36.03 43.89</td>
<td>18.02(18.02)</td>
</tr>
<tr>
<td>15</td>
<td>Taga volc.</td>
<td>Groundmass magnetite in oliv. aug. andesite</td>
<td>29</td>
<td>49.01</td>
<td>9.26(15.79)</td>
</tr>
</tbody>
</table>

* This value was calculated as 100% in total.
this table, the ferromagnetic minerals concerned here are the ferromagnetic ferrites chiefly composed of FeO, Fe₂O₃ and TiO₂.

The petrological research of the original igneous rocks has been carried out by H. Kuno chiefly from a standpoint of analysis of pyroxenes contained in them. (9) According to his opinion, No. 1 and No. 2 specimens in Table 1 are the magnetite-grains in those rocks which were cooled slowly from a low temperature, being kept in the complete equilibrium, while No. 12, No. 13, No. 14 and No. 15 specimens are the magnetite-grains in those rocks which were cooled rapidly from a relatively high temperature and No. 4, No. 5, No. 6, No. 7, No. 8, No. 9, No. 10 and No. 11 specimens are the magnetite-grains in those rocks, the character of which lies between the above two cases.

§ 3. Apparatus

The apparatus for measuring the thermal change of the magnetic susceptibility of ferromagnetic mineral grains in a weak field is the same in its principle as that used in the previous studies upon igneous rock samples. (3), (4), (6) i.e. the magnetization of specimen was measured, by a ballistic method, at any temperature in a weak magnetic field a few times as much as that of geomagnetic field.

The test specimens were packed into a silica tube 3mm in inner diameter and 10cm in length. In the case of measuring the magnetization of the ferromagnetic minerals, the demagnetizing field is practically negligible in comparison with the applied magnetic field. The electric furnace is a non-inductive and non-magnetic vacuum furnace, the air pressure in the furnace being always kept less than 1mm of mercury.

§ 4. The Susceptibility-Temperature Curves

The specimens were heated up to about 650°C in a vacuum furnace and then cooled down to the room temperature at the rate of 200°C/hour. During these processes, the change in specific magnetic susceptibility, \( \chi \), with temperature, \( T \), was measured at every 20°C by means of the above mentioned apparatus in a weak field. The applied magnetic field in these experiments is 1.35 Oe in most cases except a few examples.

The results of these measurements are illustrated in Fig. 1. As will be seen
in this figure, the mode of thermal change of susceptibility of ferromagnetic minerals is similar so much to the previous results in the cases of the igneous rocks. It seems that there are four types of the mode of the thermal change of susceptibility. The first type is that the magnitude of susceptibility has a fairly large value and in heating
Fig. 1 No. 5 $H=1.35$ Oe

Fig. 1 No. 6 $H=1.35$ Oe

Fig. 1 No. 7 $H=1.35$ Oe

Fig. 1 No. 8 $H=1.35$ Oe
process it increases gradually up to about 450°C and then decreases abruptly until the ferromagnetism disappears, and the change in cooling process follows almost the same curve reversibly. This type of change will be called here the “high-susceptibility and reversible type.” The specimens belonging to this type are No. 1 and No. 2 specimens in Fig. 1. The second is such the type as given by the \( X-T \) curve of the specimens No. 3, No. 4, No. 5, No. 6, No. 7, No. 8, No. 9, No. 10 and No. 11 in Fig. 1, in which the magnitude of susceptibility has the same order of its value as the first type and the change in heating process is similar to that of the first type but the cooling curve differs distinctively from the heating one, and besides in re-heating and re-cooling processes the susceptibility changes reversibly along the same curve as that in the previous cooling curve. This type of change will be named the “high-susceptibility and irreversible type.” The third is the type in which the magnitude of susceptibility is very small in comparison with the above-mentioned two types and it changes with temperature reversibly. This type of change will be named the “low-susceptibility and reversible type.” The specimens No. 12 and No. 13 in Fig. 1 seem to belong to this type. The forth type is that the magnitude of susceptibility has the same order of its value as the third type and it changes with temperature irreversibly. This type of change will be named the “low-susceptibility and irreversible type.” The specimens No. 14 and No. 15 in Fig. 1 seem to belong to this type. Thus, it will be summarized that there are two types “high-susceptibility” and “low-susceptibility” with respect to the magnitude of susceptibility and each of them is classified into two types “reversible” and “irreversible” with respect to the thermal change of susceptibility.

Comparing the above-mentioned classification of the \( X-T \) curves of the specimens with their petrological classification made by H. Kuno, it will be noticed that the specimens of “high-susceptibility and reversible type” and of “low-susceptibility type” of \( X-T \) curves correspond exactly to those cooled slowly from a low temperature and to those cooled rapidly from a high temperature respectively at the stage of the formation of their mother rocks, while the specimens of “high-susceptibility and irreversible type” correspond to those of petrologically intermediate type mentioned in § 2.
It seems that the correspondence mentioned above can be physically explained by taking into consideration the characteristics of the Curie-point and the magnitude of susceptibility of these mineral grains dealt with in the forthcoming paragraphs.

§ 5. The Curie-points

The Curie-point, $\theta_a$, of the specimens was defined to be the temperature at which ferromagnetism disappears apparently in the $x-T$ curve. As for the specimens of the irreversible type, there are two Curie-points, $\theta_{a1}$ and $\theta_{a2}$, corresponding to the heating and cooling curves respectively, where $\theta_{a1}$ is always higher than $\theta_{a2}$ in the present case. The values of the Curie-points thus determined are tabulated in Table 1.

The Curie-point is a physical constant of each ferromagnetic material. The Curie-point of pure magnetite, for example, has been determined to be at about $580^\circ$C. When FeO or TiO$_2$ makes a solid solution with Fe$_3$O$_4$, some amounts of decrease in the Curie-point will be observed. In order to examine the relation between the Curie-point and the chemical constitution of the specimens, $\theta_a$ of fifteen specimens were plotted against the content of TiO$_2$ in them in Fig. 2. In the case of the irreversible type specimens, $\theta_{a2}$ was taken as $\theta_a$, since it may be the more stable and consequently significant Curie-point. It will be seen in Fig. 2 that the Curie-point of the four reversible type specimens is always around $580^\circ$C and does not change with the content of TiO$_2$, while the $\theta_{a2}$ of the irreversible type specimens decreases as the content of TiO$_2$ increases.

The typical each two specimens among the two groups of the specimens of "reversible" and "irreversible" types were chemically separated into ferric and ferrous oxides and titan oxide. They are No. 1 and No. 13 in the "reversible" type specimens, and No. 11 and No. 14 in the "irreversible" type ones, the contents of FeO, Fe$_2$O$_3$ and TiO$_2$ in them being given in Table 1. In the two typical specimens of the "reversible" type, these three kinds of metallic oxides can be interpreted to compose FeO, Fe$_2$O$_3$ (magnetite) and FeO. TiO$_2$ (ilmenite) with very little excess. On the other hand, those of the two specimens of the "irreversible" type show a fair amount of FeO as an excess in the above-mentioned calculation of Norm of magnetite and ilmenite.

It may be assumed that the ferromagnetism of the "reversible" type specimens is practically that of magnetite alone, because the magnetism of ilmenite, which may simply be mixed with magnetite in this case, is far feeble compared with that of magnetite, the Curie-point being kept around $580^\circ$C regardless of the content of ilmenite.
Fig. 3 shows $\chi$–T curve of an artificial magnetite, where the Curie-point is 580°C, and the $\chi$–T curve is reversible with respect to change in temperature. This $\chi$–T curve can be considered to be the standard curve of small grain sample of almost pure magnetite. In the “irreversible” type specimens, on the other hand, FeO, Fe$_2$O$_3$, and TiO$_2$ may compose complex ferromagnetic minerals considerably different from a pure magnetite and consequently the Curie-point decreases with the increase in the apparent content of TiO$_2$.

§ 6. Magnitude of susceptibility

It will be noticed that the specimens given in Fig. 1 have a wide variety of the magnitude of $\chi$ at room temperature. For the purpose of studying the effects of impurity and grain size upon the magnitude of $\chi$, the values of $\chi$ at the room temperature were examined in connection with the content of TiO$_2$ and grain size, the result being shown in Fig. 4. That the magnitude of $\chi$ decreases as the grains become finer and as the content of TiO$_2$ increases will be fairly conceivable from the result given in this figure. However, there is also a close relation between the grain size and the TiO$_2$ content as will be seen in Fig. 4. Therefore, it becomes necessary to examine independently the dependency of the magnitude of $\chi$ on the above-mentioned two quantities. First, the dependency of $\chi$ on grain size was experimentally examined under the condition that the content of TiO$_2$ is kept constant. It has been already known that the initial permeability becomes smaller as the dimensions of the grains are diminished, since several experiments upon this subject have been reported. In the present study, the specimen No. 1, having the largest grain size, was pulverized into the smaller grains successively, and the susceptibility of their each step of grain size was measured. The results are shown in Fig. 5 together with those of the same test in an artificial magnetite and of R. Chevallier and S. Mathieu’s similar work upon hematite. These three experiments...
may be considered to show nearly the same relation between the susceptibility and grain size. As the dimensions of the particle are diminished, the relative contribution of the various energy terms to the total energy are changed. The wall energy of the boundary surfaces between domains is a surface energy and the magneto-static energy is a volume energy, being proportional to \( r^2 \) and \( r^3 \) respectively, where \( r \) is the linear dimension of representative grains.

For the small value of \( r \), then, the surface energy becomes more dominant than the volume energy. Therefore, for the purpose of minimizing the total energy, the total area of the walls must decrease at the expense of the increase of volume energy. The magnetization change in a weak field is considered to be due to the reversible wall displacement, so that it is expected theoretically that the initial permeability becomes lower as the particle becomes smaller. The experimental results shown in Fig. 5 are considered to coincide with the above-mentioned theoretical expectation.

Now, the observed magnitude of \( \chi \) of the fifteen specimens is plotted against the corresponding grain size in Fig. 6. It is obvious in this figure that the decreases of \( \chi \) with the decrease of grain size is more rapid than that expected from the result given by Fig. 5. This fact will suggest that there is some other effect controlling the magnitude of susceptibility, which may be attributed to the content of TiO₂.

It may be approximately assumed that the magnetic susceptibility, \( \chi \), of the ferrites of grain size, \( s \), can be expressed by

\[
\chi = \chi_0 \cdot g(s)
\]

where \( g(s) \) is the size factor, varying only with the size of grain, while \( \chi_0 \) denotes the susceptibility of the material having a standard dimension. The functional form of \( g(s) \) is given by the curve shown in Fig. 5. Then, the observed magnitude of \( \chi \) was normalized by means of the above relation with respect to the grain size, where the standard dimension was taken to be 410 \( \mu \) in diameter i.e. the mean diameter of

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Fig. 5

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Fig. 6
the specimen No. 1.

The values of \( \chi_0 \) thus obtained were plotted against the content of TiO\(_2\) in Fig. 7. From these results it may be summarized that the magnitude of initial susceptibility depends on the content of TiO\(_2\); the former decreasing with the increase of the latter. In the case of the reversible type specimens in which TiO\(_2\) is assumed to appear chiefly as ilmenite, the effect of TiO\(_2\) upon \( \chi_0 \) may chiefly result from the change in the content of Fe\(_3\)O\(_4\) in them. On the other hand, in the irreversible type specimens in which the Curie-point decreases with the increase of the content of TiO\(_2\) and TiO\(_2\) was assumed to be dissolved into Fe\(_3\)O\(_4\) and to be present in a complex ferromagnetic mixed-crystal, the effect of TiO\(_2\) upon \( \chi_0 \) may be chiefly due to the magnetic property peculiar to that mixed-crystal itself.

§ 7. Conclusion

From the experimental facts obtained in the present study, the chemical constitution may be considered to play an important role on the magnetic properties, especially on the Curie-point and the initial susceptibility of the ferromagnetic minerals, though they used to be called simply "magnetite" in the broad sense of the word in petrology. The grain size seems to influence the rather secondary effect upon the magnitude of susceptibility.

The reversible and irreversible properties of the thermal change of susceptibility also seem to be closely connected with the condition under which the original rock was formed from an original magma.

If such problems as mentioned above are resolved completely in the future, it could make a contribution not only to geomagnetism but also to petrology.

In concluding this paper, the writer wishes to express his sincere thanks to Dr. T. Nagata for his detailed direction throughout this study, and to Dr. H. Kuno for his kindness of putting the specimens at the writer's disposal and of offering many valuable suggestions from the petrological point of view.

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