K-Ar Ages of the Felsic Volcanism and Their Significance on Volcanostratigraphy and Kaolin Mineralization at the Hiraki Mine, Hyogo Prefecture, SW Japan

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Abstract: K-Ar ages were measured on five whole-rock samples of felsic volcanics and clays from the Hiraki mine. The results are: (1) 70.0±1.5 Ma for the underlying Kamogawa rhyolite; (2) 68.9±1.6 Ma and 69.1±1.6 Ma for the least-altered and altered rhyolitic non-welded tuff (= ore horizon), respectively; (3) 67.6±1.5 Ma for the unconformably overlying rhyolitic Hiraki welded tuff; (4) 63.8±1.5 Ma for clays from the fracture zone crosscutting the whole volcanic sequence. These ages are fairly consistent with the volcanostratigraphy in the mine area, indicating that various volcanisms and mineralization took place within a time span of a few million years in the latest Cretaceous to the earliest Tertiary Periods. The unconformity recognized, therefore, means a minor time-gap, not a long cessation of volcanism, at least in the Hiraki mine area.

1. Introduction

Hydrothermal kaolin deposit at the Hiraki mine, leading kaolin producing mine in Japan, was formed as a metasomatic product after interaction between alkali deficient peraluminous rhyolitic non-welded tuff and invading acid fluids. The stratigraphic position of this non-welded tuff, intervening the Hiraki welded tuff above and rhyolite (the Kamogawa Fm.) below, is somewhat controversial between OZAKI and MATSUURA (1988) and TANINAMI (1991) in one hand, and KO KO MYINT and WATANABE (1995) on the other. OZAKI and MATSUURA (1988) and TANINAMI (1991) placed the non-welded tuff at the lower stratigraphic position than the rhyolite, whereas KO KO MYINT and WATANABE (1995) designated that as a new unit and assigned at the upper stratigraphic position than the rhyolite lava.

Although timing of volcanism in the mine area is generally agreed to the Late Cretaceous, no age data are available for the rhyolite and the non-welded tuff, two major lithologic units, and for ores, with the exception of only two data for the youngest unit, the Hiraki welded tuff (OZAKI and MATSUURA, 1988; SHIBATA et al. 1984). In the present paper we present new age data for the whole lithologic sequence exposed in the mine area and for alteration minerals (clays) to clarify the above controversial stratigraphic sequence, as pointed out in KO KO MYINT and WATANABE (1995), and temporal relationship between mineralization and volcanic events.

2. Geologic Setting

Geological studies of the Hiraki mine and its surroundings were initiated by TANAKA et al. (1963). They assigned the Late Cretaceous acidic volcanics and pyroclastics cropping out in the area as the Arima Group with two formations: Lower — rhyolitic tuff, tuff breccia, and welded tuff; and Upper — dacitic welded tuff (TANINAMI, 1991). OZAKI and MATSUURA (1988) later correlated the Lower formation to the Kamogawa formation with four units: (in ascending order) (1) tuffaceous mud stone, sandstone and conglomerate; (2) rhyolite non-welded tuff, pumice tuff and stratified tuff; (3) rhyolite welded vitric tuff; and (4) rhyolite lava; and the Upper formation to the Hiraki Welded Tuff. The Hiraki welded tuff is subdivided into two parts: Upper Rhyolite welded vitric-crystal tuff with basal non-welded crystal vitric tuff and vitric tuff and Lower Rhyolite welded vitric-crystal tuff and tuff breccia with tuffaceous mud stone and sandstone. They defined the boundary between the Kamogawa formation and the Hiraki Welded Tuff as unconfor-
Fig. 1 (a) Location map of the Hiraki mine. (b) Geologic setting of the Hiraki mine and its environs (modified after Ozaki and Matsuura, 1988)

Very recently, the geology of the Hiraki mine area has partly been revised by Ko Ko Myint and Watanabe (1995), who proposed the new volcanic stratigraphic sequence of the Kamogawa formation in the Hiraki mine area by assuming a normal sequence and introduced the rhyolitic non-welded tuff (later altered to ore deposit) as a new lithostratigraphic unit overlying the rhyolite lava, at least in the mine area. This relation is based on the field observations which are strongly against the sequential order of Ozaki and Matsuura (1988), who described the Kamogawa formation as an overturned sequence in the Hiraki mine area. This assumption of Ozaki and Matsuura (1988) is stratigraphically irrelevant unless there is any structural or stratigraphical disturbances described, but they have never mentioned these facts in their report. Consequently, the stratigraphic sequence of the Hiraki mine area needs a serious re-establishment in accordance with its field occurrences and radiometric dates.

Moreover, Ko Ko Myint and Watanabe (1995) subdivided the non-welded tuff into three members of (1) the lower member of massive lapilli-tuff to lapillistone, (2) the middle member of an alternated sequence of medium bedded fine tuff, ash layer and lapilli-tuff, and (3) the upper member of medium, faintly bedded lapilli-tuff, in ascending order. The stratigraphic sequence established by Ko Ko Myint and Watanabe (1995) for the Hiraki mine area is shown in Fig. 2 along with radiometric ages.

Major fracture system in the mine area, trending in N70°E, is assumed to have been formed prior to the hydrothermal alteration and after emplacement of the Hiraki welded tuff. At least four large fractures, which are clearly discernible in the upper member of the non-welded tuff and fading out in the lower member probably due to pervasive alteration as well as other criss-crossing smaller ones, are possible candidates for the channel through which acid fluids incurred during the hydrothermal alteration.

3. Analytical Method

Samples collected are: (1) the fresh foot-wall rhyolite lava (sp. no. 1001); (2) the least-altered and altered non-welded tuff which formed the kaolin deposit (sp. no. 1002 and 3056); (3) the hanging-wall Hiraki welded tuff (sp. no. 1003); (4) the
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Fig. 2 Stratigraphic column of the volcanic sequence in the Hiraki mine with their respective K-Ar ages (modified after Ko Ko MYINT and WATANABE, 1995).

Table 1 Analysed sample with their respective formation and K-Ar ages. Note that determined ages are in consistent with stratigraphic sequence of Ko Ko MYINT and WATANABE (1995)

<table>
<thead>
<tr>
<th>Specimen no.</th>
<th>Material analyses</th>
<th>K (wt%)</th>
<th>$^{40}$Ar (rad/10$^6$CSP/g)</th>
<th>$^{40}$Ar% (atomic)</th>
<th>Age (Ma)</th>
<th>Remarks</th>
</tr>
</thead>
<tbody>
<tr>
<td>1014</td>
<td>Clay from fracture zone</td>
<td>2.35</td>
<td>617.3±7.8</td>
<td>18.2</td>
<td>63.8±1.5</td>
<td></td>
</tr>
<tr>
<td>1003</td>
<td>Hiraki welded tuff</td>
<td>2.89</td>
<td>772.5±8.3</td>
<td>6.2</td>
<td>67.6±1.5</td>
<td>70±3.5* 72.7±2.3#</td>
</tr>
<tr>
<td>1002</td>
<td>Non-welded tuff(least-altered)</td>
<td>1.06</td>
<td>288.8±3.6</td>
<td>17.4</td>
<td>68.9±1.5</td>
<td></td>
</tr>
<tr>
<td>3056</td>
<td>Non-welded tuff(altered)</td>
<td>1.78</td>
<td>486.6±5.8</td>
<td>10.7</td>
<td>69.1±1.6</td>
<td></td>
</tr>
<tr>
<td>1001</td>
<td>Rhyolite(Kamogawa fm.)</td>
<td>4.49</td>
<td>1244±13</td>
<td>3.5</td>
<td>70.0±1.5</td>
<td></td>
</tr>
</tbody>
</table>

* OZAKI and MATSUURA, 1988, on biotite. #SHIBATA et al. 1984, on K-feldspar.

sericite-pyrophyllite clay from the fracture zone cutting across the whole volcanic sequence in the Hiraki mine (sp. no. 1014). These samples were then crushed and sieved between mesh no. 80 and 100, and later powder specimens were used for K-Ar dating of whole rocks. To get the high sensitivity in time of formation, samples were taken from the basal part of the Hiraki welded tuff and from the uppermost part of the rhyolite. For the non-welded tuff, sample is available only from the least altered part of the middle member. In order to avoid the effect of alteration, whole-rock samples were checked carefully for their freshness before analysis. K-Ar age determination was made by use of a 30 cm radius sector-type mass spectrometer with a single collector system installed at the Okayama University of Science by the isotopic dilution method described by NAGAO et al. (1984) and ITAYA et al. (1984). The physical constant used are: $40K/K = 1.167 \times 10^{-4}$ mole/mole; $\lambda_0 = 4.962 \times 10^{-10}$/y; $\lambda_e + \lambda_\alpha = 0.581 \times 10^{-10}$/y (STEIGER and JÄGER, 1977).

4. Results and Discussion

K-Ar ages along with the K and Ar analytical data, are given in Table 1 in stratigraphical order. As shown in the table, sample from the fracture zone which includes alteration minerals, sericite
and pyrophyllite, has the youngest age of 63.8±1.5 Ma. The underlying rhyolite (the Kamogawa formation) is the oldest with 70.0±1.5 Ma, while the Hiraki welded tuff is the youngest with 67.6±1.5 Ma among the volcanic sequence. Samples of the non-welded tuff are of 69±1.6 Ma and 68.9±1.6 for altered and least-altered parts respectively. Altered specimen of non-welded tuff (no. 3056) possibly did not lose its K of igneous origin during hydrothermal alteration inasmuch as it gave out radiometric age in good agreement with that of the least-altered non-welded tuff. This fact points out that both the least-altered and altered non-welded tuff specimens represent well the fresh non-welded tuff for further geochemical purposes.

Formation of fractures, also major deformation, occurred after cessation of all volcanic events in the Hiraki mine area, since they are cutting through the Hiraki welded tuff, the youngest unit. The age of alteration minerals from the fracture zone unravels that the mineralization (hydrothermal alteration) took place about 3.8 Ma after the emplacement of the Hiraki welded tuff being formed as a cap-rock during alteration. Since there are no evidences for multiple mineralizations such as existence of mineral assemblages stable under different physico-chemical conditions, and intervening zonal array, it is assumed that the single mineralization took place in the Hiraki mine. Timing of deformation is considered as prior to mineralization, although there is a possibility of that the deformation was contemporaneous with mineralization. Exact temporal relation between deformation and mineralization cannot be justified, however.

All the above findings indicate that the whole volcanogenic rocks in the Hiraki mine area were emplaced during a relatively short duration (about 2.4 m.y.) in the latest Cretaceous to the early Tertiary Periods, and mineralization occurred after volcanic activities.

In comparison with the K-Ar ages reported by SHIBATA et al. (1984) and OZAKI and MATSUURA (1988) for the Hiraki welded tuff, the present ages are slightly young. This is probably due to the fact that their data are for the basal part of the Hiraki welded tuff, which is not exposed in the Hiraki mine area, whereas the present data are for the upper part of the Hiraki welded tuff (for details see in KO KO MYINT and WATANABE, 1995). Another possible cause is that they analyzed minerals and we used whole-rock analyses.

The relationship between clay deposits and associated igneous activities in Japan was first studied by SHIBATA and FUJI (1971), who determined the K-Ar age to be 78.7 Ma for the "Roseki" (= clayey rocks showing wax-like appearance, FUJI et al., 1976) deposit at the Yagi mine, Okayama Prefecture. SHIBATA and KAMITANI (1974) reported the K-Ar ages, ranging from 81.9 to 82.4 Ma, for the Uku "Roseki" deposit, Yamaguchi Prefecture. Recently, KITAGAWA et al. (1988) measured the K-Ar ages with the range of 87.1 to 61.4 Ma for several "Roseki" deposits in the Chugoku District and suggested a possible relation between magmatism (rhyolitic and granitic) and formation of "Roseki" deposits in that area.

These data, along with the present ones and our unpublished data (WATANABE and NISHIDO, unpublished), indicate that the "Roseki" deposits associated with so-called Late Cretaceous felsic volcanics and pyroclastics occurring in Southwest Japan were formed in the latest Cretaceous to the early Tertiary time.

Based on the mineralogical characteristics and K-Ar dating, KAMITANI (1974) and SHIBATA and KAMITANI (1974) established temporal and spatial relationship of the Uku "Roseki" deposit with the granite intrusion. However, it is pointed out here that with few exceptions (e.g., Goto mine in the Nagasaki Prefecture) such connections are still unclear.

Most of intrusive rocks of the region around the Hyogo Prefecture are of Late Cretaceous to Early Tertiary (e.g., NOZAWA, 1975) and then if there was a relation between their emplacement and formation of hydrothermal clay deposits of the same region, kaolin deposit at the Hiraki mine would be formed by infiltration metasomatism related to these igneous intrusions. Neither directly related intrusions in the nearby area, nor boron- or fluorine-bearing minerals such as those observed in the Uku deposit, is recognized in the Hiraki mine, however.

5. Conclusions

From the radiometric age data it can be concluded that the volcanics and volcanioclastics of the Hiraki mine area were formed in the following order: (from the beginning to the latest) (1) the rhyolite lava of
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References


