Properties of sedimentary clays for Karatsu ware

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Four sedimentary clays for Karatsu ware which were geologically derived from the weathered sandstone and granite were mined and used in this study. The chemical composition, mineral composition and morphology of minerals were investigated by XRF, XRD, SEM and TEM analysis. The mineral composition of normative α-quartz, feldspar and kaolin of two clays were 43.1, 4.7, and 44.3, and 52.1, 21.3, and 27.0, respectively. These clays had lower content of feldspar and higher content of kaolin than those of clays prepared from some sandstone for Karatsu ware. Average size of coarse particles in four clays was 22–28 μm and maximum particle size were 188–290 μm, and these sizes were similar to those of clays prepared from sandstone. It was suggested that the decomposition of feldspar to sericite and kaolin was promoted by the weathering and hydrolysis of sandstone or granite.

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1. Introduction

Karatsu ware was originated in the end of the 16th century in Karatsu-shi, Saga, and was developed by old Korean pottery technology such as Onggi and Punch’ong ware.1,2) Properties of Karatsu ware such as color, surface texture, sintering and mechanical strength etc. were strongly influenced by mineral composition and particle size in raw clay, glaze composition, forming, and firing process. Two types of clays which are produced from some sandstone and the weathered sedimentary clay are commercially used as raw materials for Karatsu wares. In our previous study on properties of clay prepared from ground sandstones,3) clays were composed of α-quartz, feldspar and kaolin without sericite, and the average sizes of coarse particles were 24–29 μm, and maximum particle size were 160–260 μm.

In this technical report, some properties of clays for Karatsu ware prepared from the sedimentary clay were compared with those of clays prepared from sedimentary sandstones.

2. Experimental

Four sedimentary clays were used in this study and named as A-C (Aboudani-kiln Clay), I-C (Ichinose-Kouraizin kiln Clay), H-C (Hobashira-kiln Clay), and Y-C (Yamase-kiln Clay). A-C, H-C, and I-C clays were geologically derived from the weathered sandstone, and Y-C was derived from the weathered granite. These clays were mined in the vicinity of small rivers and storage reservoirs which were located at old kiln remains operated in the end of the 16th century in Karatsu-shi, Saga Prefecture, and used as original raw samples without a traditional mill grinding and decantation. The morphology of coarse and fine particles were repeatedly treated using an ultrasonic treatment for 30 min. Ne particles were coated in water for 60 min were collected by a transmission electron microscope (TEM, model 2010, JEOL, Japan), and a transmission electron microscopy (TEM, model 2010, JEOL, Japan), respectively. From SEM images of 450 of coarse particles, the coarse particle size distribution and average particle size were calculated. Some properties of these clays were compared with those of clays prepared from sandstones.

3. Results and discussion

Figures 1 and 2 show the XRD patterns of as-prepared four clays, and TEM images of fine particles floated in water for 3 days after ultrasonic dispersion. A-C and I-C were composed of α-quartz (SiO₂), two feldspars (K₂O·Al₂O₃·6SiO₂), Na₂O·Al₂O₃·6SiO₂ and kaolin (Al₂O₃·2SiO₂·2H₂O), and H-C and Y-C were composed of α-quartz, two feldspars, kaolin and sericite (K₂O·3Al₂O₃·6SiO₂·2H₂O). In XRD pattern of Y-C, two small peaks were observed in 2θ = 6.2 and 18.6° [Fig. 1(d)]. These peaks were supposed to be one of polytype chlorite minerals,5) but the further XRD analysis for the identification of crystal phase is in progress. Y-C was only mined from the weathered granite zone in Yamase area of Karatsu-shi. From Fig. 2, four clays had smaller platy crystals (kaolinite or sericite) of 50–300nm in width, but Y-C had additionally tubular crystals of halloysite of around 200–400nm in length with platy crystals.

Table 1 shows the chemical composition of four clays. From the chemical composition and XRD result, the mineral composition of A-C and I-C was evaluated based on the normative calculation.6–7) The main mineral compositions of normative
¡quartz, feldspar, and kaolin of A-C and I-C were 43.1, 4.7, and 44.3, and 52.1, 21.3, and 27.0 mass %, respectively. These two clays had lower content of feldspar and higher content of kaolin than those of clays prepared from some sandstones. 3) Content of Al₂O₃, K₂O and Na₂O in four clays in this study were 14–20, 2.1–2.9, and 0.3–0.9 mass %, respectively. In our previous study,3) the content of Al₂O₃, K₂O and Na₂O in clays prepared from some sandstones were 13.1–17.3, 2.6–4.1, and 1.2–1.6 mass %, respectively.

The conversion of feldspar to sericite and kaolin in the Amakusa pottery stone was confirmed by a hydrothermal reaction at 180°C for 5–40 days.8) And it was reported the decomposition of K-feldspar to sericite and kaolin by the weathering and hydrolysis during a long period was progressed by the following reactions.9)

\[
\begin{align*}
(\text{I}) & \quad 3\text{K}_2\text{O}·3\text{Al}_2\text{O}_3·18\text{Si}_2\text{O}_5 + 2\text{H}_2\text{O} \\
& \quad \rightarrow 2\text{K}_2\text{O}·3\text{Al}_2\text{O}_3·6\text{Si}_2\text{O}_5·2\text{H}_2\text{O} \quad \text{(K-feldspar)} + 2\text{K}_2\text{O} + 12\text{Si}_2\text{O}_5 \\
(\text{II}) & \quad 2\text{K}_2\text{O}·3\text{Al}_2\text{O}_3·6\text{Si}_2\text{O}_5·2\text{H}_2\text{O} \quad \text{(sericite)} + 4\text{H}_2\text{O} \\
& \quad \rightarrow 3(\text{Al}_2\text{O}_3·2\text{Si}_2\text{O}_5·2\text{H}_2\text{O}) \quad \text{(kaolin)} + \text{K}_2\text{O}
\end{align*}
\]

From these reactions, XRD analysis, chemical composition and normative mineral composition in this study, it was suggested the decomposition of feldspar to sericite and kaolin was promoted by the weathering and hydrolysis of sandstone and granite for a long period.

Coarse particle in clays was α-quartz, feldspar, and kaolin of A-C and I-C were 43.1, 4.7, and 44.3, and 52.1, 21.3, and 27.0 mass %, respectively. These two clays had lower content of feldspar and higher content of kaolin than those of clays prepared from some sandstones. 3) Content of Al₂O₃, K₂O and Na₂O in four clays in this study were 14–20, 2.1–2.9, and 0.3–0.9 mass %, respectively. In our previous study,3) the content of Al₂O₃, K₂O and Na₂O in clays prepared from some sandstones were 13.1–17.3, 2.6–4.1, and 1.2–1.6 mass %, respectively.

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(\text{II}) & \quad 2\text{K}_2\text{O}·3\text{Al}_2\text{O}_3·6\text{Si}_2\text{O}_5·2\text{H}_2\text{O} \quad \text{(sericite)} + 4\text{H}_2\text{O} \\
& \quad \rightarrow 3(\text{Al}_2\text{O}_3·2\text{Si}_2\text{O}_5·2\text{H}_2\text{O}) \quad \text{(kaolin)} + \text{K}_2\text{O}
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Coarse particle in clays was α-quartz with a small amount of feldspar, and coarse particle size is one of important factor on the rough surface texture of the traditional Karatsu ware. Figures 3 and 4 show SEM images and the particle size distribution of coarse particles in four clays. Coarse particles in four clays...
had very rough and angular shape and very broad particle size distribution. Average and maximum particle size of coarse particles were 23 and 188 μm in A-C, 22 and 248 μm in H-C, 37 and 290 μm in I-C, and 28 and 198 μm in Y-C, respectively. These sizes of coarse particles were very similar to those of clays prepared from sandstone.

4. Conclusion

In this study, some properties of four sedimentary clays for Karatsu ware were investigated. The obtained results were summarized as follows:

1) Two clays (A-C and I-C) were composed of α-quartz, feldspar and kaolin, and other two clays (H-C and Y-C) were composed of α-quartz, feldspar, platy kaolin and sericite. In Y-C, fine tubular crystal of halloysite was also observed. The mineral composition of normative α-quartz, feldspar, and kaolin of A-C and I-C were 43.1, 4.7, and 44.3, and 52.1, 21.3, and 27.0, respectively. These two clays had lower content of feldspar and higher content of kaolin than those of clays prepared from some sandstone.

2) Average particle size of coarse particles in four clays were 22–28 μm, and maximum particle size were 188–290 μm and these sizes were similar to those of clays prepared from sandstone.

3) It was suggested that the decomposition of feldspar to sericite and kaolin was promoted by the weathering and hydrolysis of sandstone or granite.

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References


Fig. 4. Particle size distribution of coarse particles in the sedimentary clays.