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### Investigation of valance state of Fe ions on the firing process of celadon.

C. Kim<sup>1</sup>, J. Pee<sup>2</sup>, J. Kim<sup>2</sup>, W. Cho<sup>2</sup>, K. Kim<sup>2</sup>, C. Kim<sup>1</sup>

1. Physics, Kookmin University, Seoul, Republic of Korea; 2. Icheon Branch, Korea Institute of Ceramic Engineering and Technology, Kyeonggi-do, Republic of Korea

#### Introduction

The celadon coloration between pea green, powder green and sky green depends on the amount of the ferrous oxide (FeO) present in the glaze as well as the ion states ( $\text{Fe}^{2+}$ ,  $\text{Fe}^{3+}$ ) of iron oxide during the process [1, 2]. Therefore investigation of Fe ions during the high-temperature firing process is essential to understand the colorimetric in celadon. Mössbauer spectroscopic technique allows us to investigate samples containing very small amounts of Fe as well as their ionic conditions [3]. In this study, we have prepared celadon samples under various firing processes and investigated the change in color by varying the amount of the reducing gas. Also, we have performed and analyzed  $^{57}\text{Fe}$  Mössbauer spectroscopy measurements to understand the electronic structure and Fe ion states.

#### Experiments

Several celadon bodies with dimensions of 40\*40\*8 mm with prepared and heated to 970 °C. Glaze with specific gravity of 1.60 g/cc (55° Be) was coated on each specimen after 1st firing. As for the firing atmosphere, continuous flow of air was provided at a rate of 7 L/min. The controlled flow of Liquefied petroleum gas (LPG) was provided at different rates of 0 ~ 3.5 L/min in order to generate CO gas as for a reducing agent. LPG was introduced at 980 °C for 30 minutes and after that, the temperature was increased up to 1250 °C. The crystal structure of the sample was examined by XRD. The samples of celadon from various firing processes were measured using the Handy NR-3000 colorimeter. An oxygen/nitrogen analyzer was used to investigate the amount of oxygen remaining within the glaze. The Mössbauer spectra were recorded with a fixed absorber and a moving source by using a conventional spectrometer of the electromechanical type with a  $^{57}\text{Co}$  source in a Rh matrix.

#### Results and discussion

According to the chromaticity ( $L^*$ ,  $a^*$ ,  $b^*$  values) results, the color changes from Yellow-Red (YR) to Green-Yellow (GY) with increasing brightness as the amount of LPG increases. As the amount of LPG flow gradually increased, the value of  $a^*$  changed from 1.6 to -18.9 and the value of  $b^*$  changed from 33.1 to -10.6. Figure 1 shows the Mössbauer spectra for different firing processes (LPG = 0 ~ 3.5 L/min) at room temperature. As shown in Fig. 1, all samples did not exhibit any magnetic behaviors.

Figure 2 shows  $\text{Fe}^{2+}/\text{Fe}^{3+}$  values at room temperature. For different firing processes therefore we have analyzed Mössbauer spectra with two Fe sites showing paramagnetic behavior. From the Mössbauer analysis, the  $\text{Fe}^{2+}/\text{Fe}^{3+}$  value of the sample with fired glaze in the oxidizing atmosphere (LPG = 0 L/min) was 0.1, while that of the sample with fired glaze in the reducing atmosphere (LPG = 3.5 L/min) was 4.26. When the amount of LPG increased, the area ratio of  $\text{Fe}^{3+}$  ion decreased, while that of  $\text{Fe}^{2+}$  ion increased. Increasing flow of LPG implies the increase in reduction gas (CO). Therefore a large number of Fe ions tend to change from  $\text{Fe}^{3+}$  ions to  $\text{Fe}^{2+}$  ions. This is consistent with the change in chromaticity from YR to GY. Especially, the rapid change in the amount of LPG flow at 1.5 L/min confirms the change in the  $\text{Fe}^{2+}/\text{Fe}^{3+}$  ratio.

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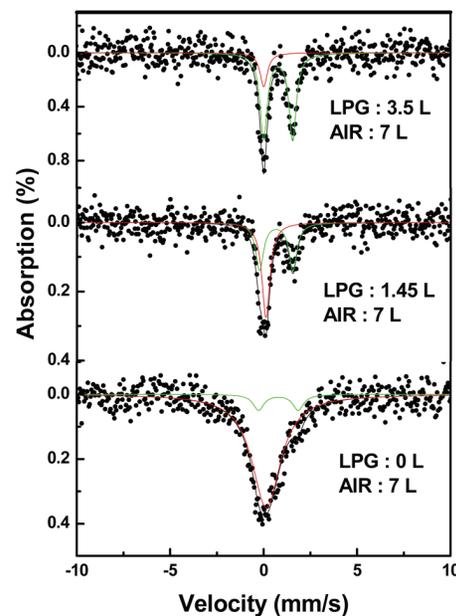


Fig. 1. The Mössbauer spectra according to the firing process (LPG = 0 ~ 3.5 L/min) at room temperature.

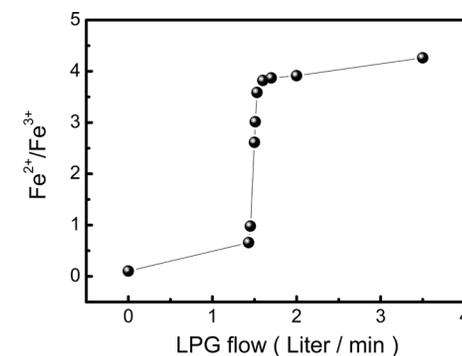


Fig. 2. The firing process (LPG = 0 ~ 3.5 L/min) dependences of  $\text{Fe}^{2+}/\text{Fe}^{3+}$  values at room temperature.