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学 位 論 文 要 旨
Dissertation Summary

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論 文 名 ： Viscosity, Density, and Surface Tension of Gasified Coal and Synthesized Slag Melts for Next-Generation IGCC

(Dissertation Title)

In this century, integrated gasification combined cycle (IGCC) as coal-based power plant technology is expected to grow by over 40% in the next 20 years in operation worldwide due to high efficiency and environmental compliance. Furthermore, the gasification process applied to a variety of coal grades at high temperatures is expected to flow down coal slag melts stably and continuously into the slag bath as a gasified coal. However, limited studies on the measurements of the coal slag melt properties have been reported because experimentally difficult to carry out, expensive, and time consuming. The slag melts properties are essential for engineering background and a scientific point of view. The objectives of this study are to investigate viscosity, density, and surface tension of gasified coal and synthesized slag melts at high temperatures in the range of 1300 to 1650 °C. Emphases have been placed on the following points. The compositional and temperature dependences of gasified coal and synthesized slag melt properties were studied. The composition parameter to predict the viscosity, density and surface tension was proposed based on chemical composition.

This thesis consists of five chapters as follows:

In chapter 1, the background, previous studies, and purposes of the present study were summarized.

In chapter 2, the viscosities of gasified coal and synthesized slag melts were measured in air by a rotation cylinder method. The reproducibility of the measurement at high temperatures was evaluated for the viscosities of $50CaO–50SiO_2$ (mol%) and deviation was estimated to be 1.5% compared with the viscosity data of slag melts in a published literature. The viscosity decreased monotonically with increasing temperature for all samples. The main components of gasified coal slags are SiO_2, Al_2O_3, CaO, MgO, FeO and Fe_2O_3. Slags consisting of the main components with systematic composition variations were synthesized and evaluated for comparison. The experimental results of chelate titration analysis revealed that the percentages of Fe^{3+} in coal and synthesized slag samples were 47–56% and 68–80% of total Fe, respectively. The viscosity of AD slag melt with relatively-high Fe_2O_3 (~8 mol%) and low SiO_2 (~40 mol%) contents rapidly increased with decreasing temperature from 2 Pa·s

at 1350 °C to ~10 Pa·s at 1300 °C presumably due to crystallization. The viscosity of gasified coal slag melts increased with increasing contents of Al_2O_3 and SiO_2 and decreased with increasing contents of CaO, MgO, FeO, and Fe_2O_3. The roles of these main components on melt viscosity were classified into two types of network former (NWF) and modifier (NWM). Two types of amphoteric oxides: Al_2O_3 and Fe_2O_3 play different respective roles of NWF and NWM on viscosity for gasified and synthesized slag melts in CaO–MgO–Al_2O_3–FeO–Fe_2O_3–SiO_2 system. A composition parameter relating to the total contents of NWF and NWM is proposed to optimize the viscosity of slag melts at high temperatures, compared to moderate viscosities of 5–15 Pa·s for the entrained-flow gasifier of IGCC.

In chapter 3, the densities of gasified coal and synthesized slag melts were measured in air by an Archimedean double-bob method. The reproducibility of the measurement at high temperatures was evaluated for the densities of 40Na_2O–60SiO_2 (mol%) and deviation was estimated to be 0.1% compared with the density data of slag melts in a published literature. The density decreased linearly with increasing temperature for all melt samples. The density of gasified and synthesized coal slag melts was found to decrease with increasing Al_2O_3 and SiO_2 contents and to increase with increasing Fe_2O_3 and FeO contents. The molar volume and the coefficient of volume expansion, which were calculated from measured density values, increased monotonically with increasing Al_2O_3 content for CaO–Al_2O_3–SiO_2 (CAS) synthesized samples with 40 mol% SiO_2. The molar volume for CaO–FeO–Fe_2O_3–SiO_2 (CFS) synthesized samples with 60 mol% SiO_2 increased with Fe_2O_3 addition and exhibited discontinuous change at ~7.5 mol% Fe_2O_3. The coefficient of volume expansion in the same CFS series decreased with increasing Fe_2O_3 content showing a minimum value at ~7 mol% Fe_2O_3 and increased in higher Fe_2O_3 content. A composition parameter calculated from chemical composition was proposed to predict the density of coal slag melts. The relationship between measured density and the composition parameter was analyzed.

In chapter 4, the surface tensions of synthesized slag melts were measured in air by a maximum bubble pressure method. Ar gas is blown to the slag melts through Pt–13%Rh capillary tube for produced bubbles. The reproducibility of the measurement at high temperatures was evaluated for the surface tension of 40CaO–20Al_2O_3–40SiO_2 (mol%) and deviation was estimated to be 1.87% compared with the surface tension data of slag melts in a published literature. The synthesized slag melts compositions were chosen based on the main components of the gasified coal slags. It was found that the surface tension of CAS slag melts increases with increasing temperature, on the contrary, surface tension of CFS slag melts decreases with increasing temperature. The surface tension decreases with addition of Al_2O_3 content in CAS slag melts at fixed SiO_2 content. It also found to decrease with increasing SiO_2 content in CAS and CFS slag melts at constant Al_2O_3 and Fe_2O_3 contents. The surface tension monotonically decreases when Al_2O_3 is replaced by Fe_2O_3 in CFS slag melts. An empirical composition parameter at temperatures of 1400, 1500 and 1600 °C is proposed to predict the surface tension from the corresponding chemical composition of measured synthesized slags.

In chapter 5, the general conclusions of this study were summarized.