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Viscosity measurement and prediction of gasified and synthesized coal slag melts

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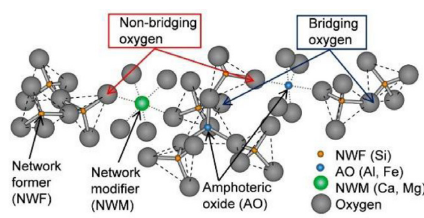
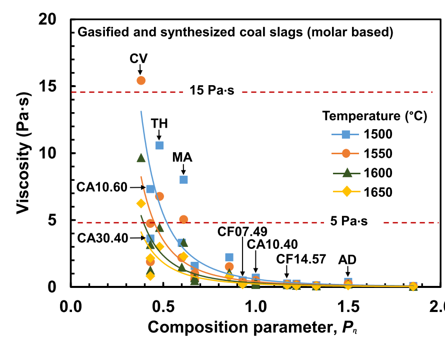
HIGHLIGHTS

- Viscosity of gasified and synthesized coal slag melts was measured in air.
- Composition dependence of viscosity was studied.
- Additions of Al_2O_3 and Fe_2O_3 exhibit different effects on viscosity.
- A composition parameter was proposed based on measured viscosity of slag melts.

GRAPHICAL ABSTRACT





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ABSTRACT

The viscosity of gasified and synthesized coal slag melts for homogeneous liquid state was measured in air by rotation cylinder method with the viscosity range of 0.2–15 Pa·s in the temperature range 1300–1650 °C. The viscosity decreased monotonically with increasing temperature for all samples. 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 are classified into 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 parameter relating to the total contents of NWF and NWM is proposed to predict the viscosity of slag melts at high temperatures from chemical composition.

1. Introduction

The Integrated gasification combined cycle (IGCC) of the Central Research Institute of Electric Power Industry (CRIEPI) commissioned in a project of New Energy and Industrial Technology Development Organization (NEDO), Japan has been designed to produce electricity with CO_2 capture [1,2]. The organic compounds of coals are converted to synthesis gas while the inorganic compounds become coal ash during the gasification process. The slag melt due to the coal mineral matters is discharged through a hole under the entrained-gasifier by quenching into water as a gasified coal slag [3,4]. Viscosity is one of the key properties to determine the flow behavior of slag melts [5].

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The flow behavior affects the performance of continuous operation of the entrained-flow gasifier [6,7]. The viscosity of slag melts should have typically adequate values of 5–15 Pa·s for entrained-flow gasifiers at tapping temperature with good flow [8,9]. The influence of slag melt viscosity varies as a function of gasifier design, operating temperature, atmosphere, and the chemical composition of coal ash slag [9,10]. Large numbers of researches have studied for the composition and temperature dependences of coal slag viscosity [11,12].

The slags formed during the coal gasification process mainly contain SiO_2, Al_2O_3, CaO, MgO, FeO and Fe_2O_3 with small amounts of Na_2O, K_2O, TiO_2, P_2O_5, SrO, MnO, and other compounds [8,13]. In the main components, a previous study reported that roles of Al_2O_3 and Fe_2O_3 as amphoteric oxides depend on slag composition and melting conditions [14].

According to these scientific and engineering backgrounds, the objectives of this research are (i) to establish a rotation cylinder method to measure viscosity of gasified and synthesized coal slag melts for homogeneous liquid state in silicate systems containing CaO or MgO and Al_2O_3 and/or FeO/Fe_2O_3, (ii) to study the composition and temperature dependences of viscosity, (iii) to discuss the effect of Al_2O_3 and Fe_2O_3 on viscosity, and (iv) to propose a parameter to predict the viscosity of coal slag melts based on chemical composition.

2. Materials and methods

2.1. Sample preparation

Coal slag samples denoted as CV (Coal Valley), TH (Tanito Harum), MA (Malinau), and AD (Adaro) were provided by the CRIEPI of Japan. Coal slag samples were chosen to represent coals with various compositions from different countries and coal ranks produced from the gasification runs instead of raw coals. Except for AD slag, the compositions of coal slags had been determined in our previous study [14] as shown in Table 1. The contents of coal slags only show major components of gasified coal slags without including minor components. The valence state of iron oxide was evaluated using chelate titration method [15] in quenched samples.

Table 1 also shows that the batches of synthesized slag samples are denoted into three composition series nominally at 40, 50, and 60 mol% SiO_2 contents with various contents of Al_2O_3, CaO, MgO, and Fe_2O_3 for comparison. As for synthesized slags, we selected similar composition ranges appeared in the coal slags for Al_2O_3, CaO, and Fe_2O_3. A composition including MgO for a synthesized slag named MA10.60 is also selected in substitution for CaO. The synthesized slag samples were prepared from a mixture of SiO_2, Al_2O_3, $CaCO_3$, MgO, and Fe_2O_3 powders with 99.99% purity as raw materials. The mixtures were melted in air using a Pt-30wt% Rh crucible between 1350 and 1650 °C for about two hours depending on their homogenous melting temperatures. The synthesized slag melt was poured onto iron mold, cooled down to room temperature and then crushed for viscosity measurements. The fraction of Fe^{3+}/Fe_{tot} was also analyzed experimentally in quenched samples after viscosity measurements using the chelate titration method as shown in Table 1.

In the case of AD coal slag, the chelate titration method was not performed due to the difficulty in dissolution of AD powder sample. Thus, the iron oxide in the AD coal slag was assumed to be Fe_2O_3. A previous study [16] reported a similar result that all iron exists as ferric iron in a coal slag consisting mainly of SiO_2, Al_2O_3, CaO, and MgO with 5 mol%Fe_2O_3 under the melting below 1400 °C in air.

2.2. Viscosity measurements of slag melts

The slag viscosity was measured in air as the temperature dropped from 1650 to 1300 °C using a rotating cylinder method. The measuring temperature of viscosity was selected for homogeneous melting state. The thermodynamic software package FactSage [17] was used in this study to predict liquidus temperature

Table 1
Composition of major component of gasified coal slags and synthesized slags for viscosity measurements.

Series	Sample	Fe^{3+}/Fe_{tot} [a]	Molar ratio (mass ratio) or mol% (mass%) [b]												T_L or T_{hm} [d] (°C)	Basicity parameter [e]
			SiO_2		Al_2O_3		CaO		MgO		FeO [c]		Fe_2O_3 [c]			
Coal slag	Coal Valley (CV)	0.56	61.5	(54.8)	13.9	(21.1)	14.2	(11.9)	3.6	(2.1)	2.2	(2.3)	1.4	(3.3)	1500	0.28
	Tanito Harum (TH)	0.47	57.8	(49.8)	16.0	(23.4)	9.2	(7.4)	4.8	(2.8)	5.8	(6.0)	2.6	(6.0)	1450	0.30
	Malinau (MA)	0.52	54.6	(46.2)	15.0	(21.6)	8.0	(6.3)	6.3	(3.6)	7.1	(7.2)	3.9	(8.7)	1450	0.36
	Adaro (AD)	–	39.0	(34.3)	10.0	(14.9)	24.5	(20.1)	16.4	(9.7)	0.0	(0.0)	8.0 [f]	(18.7)	1250	1.00
(60-x)CaO–xA_2O_3–$40SiO_2$	CA10.40	–	40.0	(38.6)	10.0	(16.4)	50.0	(45.0)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1351	1.00
(A = Al and/or Fe)	CA20.40	–	40.0	(35.9)	20.0	(30.5)	40.0	(33.6)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1358	0.67
	CA30.40	–	40.0	(33.6)	30.0	(42.8)	30.0	(23.5)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1509	0.43
	CF08.39	0.80	39.2	(35.5)	0.0	(0.0)	49.0	(41.4)	0.0	(0.0)	4.0	(4.3)	7.8	(18.8)	1362	0.55
	CAF15.11.39	0.76	38.6	(30.2)	14.5	(19.3)	29.0	(21.2)	0.0	(0.0)	6.9	(6.5)	11.0	(22.9)	1428	1.55
(50-x)CaO–xA_2O_3–$50SiO_2$	CA00.50	–	50.0	(51.7)	0.0	(0.0)	50.0	(48.3)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1541	0.88
(A = Al or Fe)	CA12.50	–	50.0	(47.1)	12.5	(20.0)	37.5	(33.0)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1295	1.00
	CF07.49	0.69	48.5	(44.2)	0.0	(0.0)	38.8	(33.0)	0.0	(0.0)	6.1	(6.6)	6.7	(16.2)	1202	0.60
(40-x)RO–xA_2O_3–$60SiO_2$	CA00.60	–	60.0	(61.6)	0.0	(0.0)	40.0	(38.4)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1457	1.06
(R = Ca or Mg)	CA10.60	–	60.0	(57.2)	10.0	(16.2)	30.0	(26.7)	0.0	(0.0)	0.0	(0.0)	0.0	(0.0)	1317	0.70
(A = Al or Fe)	MA10.60	–	60.0	(61.8)	10.0	(17.5)	0.0	(0.0)	30.0	(20.7)	0.0	(0.0)	0.0	(0.0)	1363	0.43
	CF07.58	0.71	58.3	(52.7)	0.0	(0.0)	29.2	(24.6)	0.0	(0.0)	5.6	(6.1)	6.9	(16.6)	1258	0.43
	CF14.57	0.74	57.1	(46.0)	0.0	(0.0)	19.0	(14.3)	0.0	(0.0)	9.8	(9.4)	14.1	(30.2)	1308	0.72

[a] The fraction of Fe^{3+}/Fe_{tot} was analyzed two or three times for quenched samples using chelate titration method.
[b] Molar ratio (mass ratio) for gasified coal slags was obtained based on the contents of major components only. The total amount of minor components was less than 2.1 mol% (1.9 mass%). Synthesized slags consist of the major components only and their compositions are indicated.
[c] The contents of FeO and Fe_2O_3 were calculated from analyzed Fe^{3+}/Fe_{tot}.
[d] Liquidus temperatures T_L for synthesized slags were obtained using FactSage data bank. Homogeneous melting temperatures T_{hm} for gasified coal slags were determined experimentally by hot thermocouple (HTC) method.
[e] Basicity parameter (base/acid ratio) without including Fe_2O_3: $(CaO + MgO + FeO)/(Al_2O_3 + SiO_2)$ in molar ratio.
[f] All iron oxide was assumed to Fe_2O_3 in Adaro coal slag.