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Surface tension measurements of coal ash slags under reducing conditions at elevated pressures

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ABSTRACT

The aim of limiting the amount of CO_2 that is released together with other exhaust gases from power plants can be reached by technologies allowing for a systematic separation of this greenhouse gas. One such technology is the integrated gasification combined cycle power plant which makes use of a coal gasification step. For the gasification involving temperatures far higher than in typical pulverised combustion chambers, ash contained in the fuel is liquefied (slag) and must be removed from the cycle to guarantee safe operation of downstream equipment. To keep the efficiency of the power plant as high as possible, hot gas cleaning facilities are most desirable for this purpose. The design of these installations necessitates knowledge about thermophysical properties of coal ash slags, especially in reducing, pressurised atmospheres. In this work, the surface tension of 15 coal ash slags was measured in argon hydrogen gas of up to 10 bar absolute pressure according to the sessile drop method. Compared to experiments at 1 bar, surface tension values up to 42% lower were found on applying pressure. Additionally, shifts in the melting temperature interval of the ashes due to increased pressure were observed. The surface tension values obtained in pressurised atmospheres ranged from 270 to 490 mN/m with respect to temperature intervals where almost no data scattering occured.

1. Introduction

Due to increasing concern about CO_2 emissions from power plants in the context of global warming, technologies allowing for a cleaner production of electricity from coal need to be developed. One such technology is referred to as integrated gasification combined cycle (IGCC) in the literature [1–5]. This power plant process unifies a coal gasification facility and the well-established combined cycle technology. The major advantage of gasifying coal instead of burning it, lies in the fact that a CO_2 removal unit can be installed downstream of the gasification vessel [6]. Therefore, an unrestricted release of this greenhouse gas into atmosphere is avoidable and the CO_2 can instead be stored in a safe deposit. In order to profit from this technology, one has to deal successfully with challenging process parameters in the gasification reactor, namely temperatures of up to 1800 °C and pressures of up to 100 bar [5]. Such conditions lead to a liquefaction of the ash contained in the gasified coal. The corresponding slags are highly corrosive, so that installations following the gasifier have to be prevented from contact with them.

Aiming at the removal of slag particles entrained in the gasifier's output gas (syngas), water quench installations are used nowadays [7]. This approach worsens the overall efficiency of IGCC, so that future efforts must be aimed at developing hot gas cleaning facilities [8]. Making use of what already worked successfully in pressurised pulverised coal combustion (PPCC) [9], ceramic balls acting as a filter should be integrated into the flow path of syngas. A removal of slags at the bottom of the cleaning installation can thus be realised at unaltered temperatures guaranteeing a high efficiency.

In order to correctly construct the described hot gas cleaning facility, thermophysical data on coal ash slags under gasification conditions (reducing atmosphere) need to be known. Especially for the surface tension of coal ash slags in *pressurised* atmospheres, to the best of the authors' knowledge no data are given in the literature. Various researchers investigated the surface tension of such substances at atmospheric pressure and even in reducing environments, but high pressures in the gasification vessel are thought to influence thermophysical properties by accelerating chemical processes. Therefore, the surface tension of coal ash slags also needs to be studied at elevated pressures. If the influence of pressure on the surface tension is well-understood, data from atmospheric pressure experiments can possibly be exploited to predict the surface tension change in gasification reactors.

Besides earlier experiments conducted by the authors at atmospheric pressure [10,11], Falcone [12,13] reports surface tension values for four different American coal slags. The author exposed

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ash pellets to an inert gas atmosphere and measured the surface tension of the corresponding molten slag by means of the sessile drop method. The experimental results range from 364 to 1489 mN/m for temperatures between 1225 and 1285 °C. Vitreous carbon is mentioned as suitable substrate material because of its non-wetting character.

For almost the same coals Schobert [14] lists the measurement values of Miller and Kalmanovitch [15] which suggest a lower slag surface tension in oxidising atmospheres compared to reducing ones. In contrast, Schobert also provides data indicating a surface tension increase when changing from reducing to oxidising conditions. For a reducing environment, surface tensions between 377 and 615 mN/m were published for the temperature range from 1234 to 1275 °C.

Furthermore, Raask [16] measured a surface tension of 320 mN/m for a coal ash slag in the temperature interval from 1300 to 1400 °C. Another data point for a gasifier slag can be found in the article by Mills and Rhine [17] who observed a surface tension value of 430 mN/m at 1350 °C. Just like Mills and Rhine, Nowok et al. [18,19] also employed vitreous carbons as substrate material in sessile drop experiments and found surface tension values ranging from 190 to 960 mN/m for different temperatures and varying gas atmospheres.

In addition to concentrating on real ash slags, model systems must be investigated in a systematic fashion to understand the influence of individual ash components on surface tension [12] and to judge the suitability of specific coals for hot gas cleaning processes. Studying model systems will pave the way for enlarging property databases and for more coals being used in *pressurised* hot gas cleaning facilities by the employment of additives.

2. Materials and methods

As an extension of a typical sessile drop apparatus described frequently in the literature [18,20–24], a furnace designed for gas atmospheres of up to 20 bar absolute pressure was mounted in front of a CCD camera attached to a zoom lens. In spite of the design pressure, current experiments were only conducted at 5 and 10 bar absolute pressure. The results could be compared to surface tension data obtained earlier at atmospheric pressure.

Ash samples from coals (all ashed at 815 °C in air, except for the HKT sample which was ashed at 450 °C in air) mined in four different countries were pressed into pellets (5 mm in diameter and approximately 5 mm in height) and positioned in the furnace to be melted to liquid drops. In addition to the ashes (powder form), three samples from an experimental gasification facility (glassy pieces) in Germany were also investigated (S1-1, S1-2 and S1-4). For confidentiality reasons, the authors were not provided with the corresponding gasifier conditions by the industrial project partners. The compositions of the studied materials can be found in Table 1. All ST-D samples originate from deep coal mines in Germany, whereas the ST-N coals were mined in Norway (Spitsbergen). ST-P-1 was extracted in Poland. In addition to these black coal samples, three brown coal ashes from opencast mines in Germany were included in the experimental schedule (HKT, K2-1 and K3-1). Prior to performing experiments, all samples were stored in closed plastic containers in air atmosphere at ambient temperature.

In order to calculate the surface tension of the materials under investigation, three different computer algorithms were used to analyse drop images. Those photos were taken in intervals of 1 °C while constantly heating the furnace at a rate of 2 °C/min. As one of the computer codes permanently produced heavily scattering surface tension results, the further discussion will be limited to the “ADSA” and the “LBADSA” program in this article. ADSA relies on numerically integrating the *exact* Young–Laplace equation prior to fitting solution curves to the drop profile found in an experimental image and is frequently cited in the literature [20,24–32]. LBADSA instead uses an *approximate* solution of the Young–Laplace equation and fits it to the drop contour by means of active contours or snakes [33–35].

As it was previously ascertained that the results of both codes are in good agreement with each other (mostly an offset in the surface tension can be observed), ADSA data were always weighted by 60%, whereas LBADSA got a weight of 40% when fitting functions were applied or averages were calculated. All data within this article that are shown as lines (solid, dashed, dotted, etc.) represent such fitting functions. In contrast, symbols in diagrams refer to a weighted average of the results by ADSA and LBADSA. Whenever surface tensions were calculated from the capillary constant $c = \frac{\Delta\rho \cdot g}{\sigma}$ outputted by the algorithms, the density of the surrounding gas phase in $\Delta\rho = \rho_{\text{liquid}} - \rho_{\text{gas}}$ was neglected (g refers to gravitational acceleration and σ to surface tension). Even elevated pressures do not lead to the gas density ρ_{gas} being significant compared to the slag density ρ_{liquid} derived from drop volume and sample mass.

A more detailed description of the measurement facility, the image analysis algorithms employed and the sample preparation can be found in an earlier publication by the same authors [11].

Because of the literature citing graphite [16,21,36,37] and vitreous carbon [12,13,15,17–19] as suitable substrates in the context of sessile drop experiments on coal ash slags, three different types of such materials were investigated for their wetting behaviour. Namely these are the graphite 5890PT manufactured by Carbone Lorraine (France), the graphite FE 479 by Schunk (Germany) and

Table 1
Ash/slag compositions in equivalent oxides (weight percentages).

	Al_2O_3	BaO	CaO	Fe_2O_3	K_2O	MgO	Mn_2O_3	Na_2O	SiO_2	TiO_2
ST-D-1	26.26	0.09	2.94	10.72	2.53	2.32	0.16	1.04	43.86	1.00
ST-D-2	20.03	0.21	7.00	9.58	2.17	3.98	0.23	1.35	40.43	0.80
ST-D-3	26.83	0.12	4.06	8.58	3.37	2.32	0.09	1.24	42.79	0.98
ST-D-4	25.70	0.11	3.50	7.58	3.49	2.82	0.12	0.80	47.92	0.95
ST-D-5	23.43	0.18	3.64	7.44	3.98	2.82	0.11	1.00	49.85	0.92
ST-D-6	29.29	0.19	3.50	20.45	1.69	1.53	0.13	2.02	39.15	0.53
ST-N-1	13.42	0.28	11.05	9.58	1.45	3.81	0.04	2.83	37.22	0.85
ST-N-2	20.22	0.31	9.74	10.31	2.78	2.67	0.02	5.47	41.93	1.03
ST-P-1	24.00	0.25	6.80	7.99	2.96	4.26	0.14	2.16	46.85	1.14
HKT	17.01	0.12	12.03	2.15	0.63	4.98	0.04	2.29	44.50	1.10
K2-1	2.12	0.39	24.07	9.58	0.15	7.84	0.16	1.90	38.08	0.43
K3-1	3.10	0.09	7.07	13.07	0.55	2.26	0.07	0.08	56.91	0.23
S1-1	3.02	0.15	19.45	8.02	0.27	5.64	0.14	1.52	60.54	0.30
S1-2	24.75	0.67	12.62	4.86	0.93	2.34	0.08	1.78	48.14	0.48
S1-4	12.28	0.34	15.11	7.62	1.49	4.31	0.08	4.77	52.41	0.52