OXY-FUEL COMBUSTION OF COAL UNDER HIGHER PRESSURE AND LOW OXYGEN CONCENTRATION

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Abstract – Oxy-fuel combustion (OFC) is one of the way to efficient reduce CO_2 emission to the atmosphere. The development of oxy-fuel combustion is currently inhibited due to few important issues. The current politics and public opinion is rather critical for OFC. Moreover, the part of scientific community criticises this technology due to the cost and huge energy penalty (Stanger, Wall et al. 2015; Wu, Yang et al. 2016). However, they are still the projects where efforts lead to obtain demonstration scale plants of oxyfuel technology. Another important barrier to full scale commercialization of OFC is oxygen demand. It was estimated by Chorowski et al. (Chorowski, Gizicki et al. 2012) that 1 MW_e power requires 18 000 kg O_2 per day. From another side, OFC provides many benefits, mainly related to lower gaseous pollutants emission. These effects can be enhanced by the increase of pressure inside combustion chamber. The aim of this paper is to show the benefits of the pressurized OFC. The results of combustion at low oxygen concentration (15 v.%) OFC under elevated pressure (up to 6 bar) are presented. The experiments were carried out using autothermal bubbling fluidized bed combustor (diameter of 0.075 m, height of 1 m and static bed height of 0.25 m). Ziemowit hard coal was used as fuel. The relevant results of the investigations consist the effect of combustion stabilization under higher pressure, very low NO emission (below 30 mg/m_n^3 (STP) at 6 v.% O_2) and the decrease of CO emission.

INTRODUCTION

Oxy-fuel combustion (OFC) is one of the way to efficient reduce CO_2 emission to the atmosphere. The development of oxy-fuel combustion is currently inhibited due to few important issues. The current politics and public opinion is rather critical for OFC. Moreover, the part of scientific community criticises this technology due to the cost and huge energy penalty (Stanger, Wall et al. 2015; Wu, Yang et al. 2016). However, they are still the projects where efforts lead to obtain demonstration scale plants of oxyfuel technology. Another important barrier to full scale commercialization of OFC is oxygen demand. It was estimated by Chorowski et al. (Chorowski, Gizicki et al. 2012) that 1 MW_e power requires 18 000 kg O_2 per day. From another side, OFC provides many benefits, mainly related to lower gaseous pollutants emission. These effects can be enhanced by the increase of pressure inside combustion chamber. It is known that higher pressure in the combustion chamber creates a preferential conditions to inhibit NO_x creation during combustion process (Lasek, Janusz et al. 2013). Pressurized oxy-fuel combustion (POFC) is also suggested as a one technology within clean coal technologies (CCT). Due to potentially higher power generation efficiency, it was suggested that pressurized oxy-fuel combustion systems had the potential of better performance over conventional atmospheric oxy-fuel combustion power cycles (Hong, Chaudhry et al. 2009)

The aim of this paper is to show the emission characteristics of the pressurized OFC. The results of combustion at low oxygen concentration (15 v.%) OFC under elevated pressure (up to 6 bar) are presented. Ash-issues were analyzed using characteristic indexes to determine the risk of slagging and fouling.

EXPERIMENTAL

All fuel analyses (proximate, ultimate, low heating value, LHV) were carried out according to the procedures. Samples of biomass were grinded to the size <0.425 mm. The ash of the biomass sample was achieved by the combustion of sample portion in electric heated furnace at temperature of 600°C. Elemental analysis was performed on a LECO TrueSpec (LECO, USA) CHN and a LECO SC 632 (LECO, USA). LHV was measured using a LECO AC500 (LECO, USA).

Experiments were carried out using a laboratory-scale fluidized bed (bubbling regime), auto-thermal combustor as described previously (Lasek, Janusz et al. 2013). The main component of the experimental setup was a fluidized bed combustor (bubbling regime). The reactor consisted of a stainless steel tube with a reactor diameter of 0.075 m, height of 1 m and static bed height of 0.25 m. The flue gas was continuously analyzed for CO_2 (0-45 v%), CO (0-6000 ppmv), H_2O (0-30 v.%), NO (0-1000 ppmv), N_2O (0-200 ppmv), NO_2 (0-200 ppmv) and HCl (0-100 ppmv) using an FTIR analyzer (GASMET DX4000), and O_2 (inlet and outlet) was measured by a zirconium sensor analyzer (AMS Analysen). The measurement uncertainty of presented analyzers was below 2 % of the total measuring range. Silica sand (diameter of 0.7-1 mm) was used as the bed material. Temperature of freeboard (inside combustion chamber, $T_{freeboard}$) was measured using K-type thermocouple (with shells) in point at 320 mm from the grid. According to the certificate provided by the thermocouple producer, the measurement uncertainty was below 5°C. A point of the pressure measurement was 1110 mm above the grid (top zone) and 120 mm below the grid.

Ziemowit hard coal was used as fuel in this research. Ultimate and proximate analysis of Ziemowit coal is presented in Table 1.

Table 1. Ultimate and proximate analysis of Ziemowit coal

Proximate analysis (wt.%, air-dry state)	
Moisture	3.6
Ash	24.1
Volatile matter	29.2
Fixed carbon	43.1
LHV (kJ kg^{-1})	21449
Ultimate Analysis (wt%, air-dry state)	
S(total)	1.31
S(c)	1.04
C	56.2
H	3.76
N	0.85
O (calculated)	10.45
Other compounds (wt%, air-dry state)	
P	0.017
Cl	0.431
c- combustible matter	

RESULTS AND DISCUSSION

Emission issues

Fig. 1. shows the example of measured flue gas composition in function of time. It can be noticed that the inlet O_2 concentration was maintained at level of ~15 vol.%. The average values of measured compounds were determined and presented in Table 2. Combustion under atmospheric pressure was unstable. It was confirmed by the high measured values of CO and CH_4 volume fractions. Fluctuations of measured parameters was in wide range (e.g. for CO at high deviation of 2870 mg/m_n^3). Combustion was stabilized when pressure was increased. CO concentration decreased and temperature inside combustion chamber was higher. The relatively high emission of HCl was caused by the chlorine content in the fuel (see Table 1). It can be noticed that NO emission was very low compared to the pressurized oxy-fuel combustion at higher O_2 concentration inlet. It was observed before that NO emission for O_2≈20 vol.% was higher than 100 mg/m_n^3 (STP, referred to 6% O_2) (Lasek, Janusz et al. 2013). Both parameters, i.e. higher pressure inside combustion chamber and low O_2 concentration were preferential to decrease NO_x emission. It has been suggested that pressurized combustion creates favorable conditions for the NO-char reaction (NO+C→CO+1/2N_2), leading to low NO emissions (Croiset, Heurtebise et al. 1998; Lasek, Janusz et al. 2013). Moreover, higher pressure

was also preferential to the decrease of CO emission. The decrease of N_2O emission is rather caused by increase of temperature inside combustion chamber and destruction of N_2O (Leckner 1998). The nitrous oxide emissions are sensitive to temperature and pressure rather enhanced N_2O formation (Svoboda and Pohořelý 2004). Fig. 2 shows the comparison of NO and CO emissions during low-oxygen combustion of Ziemowit hard coal. Grey marked box concerns the stable combustion at low-oxygen concentration.

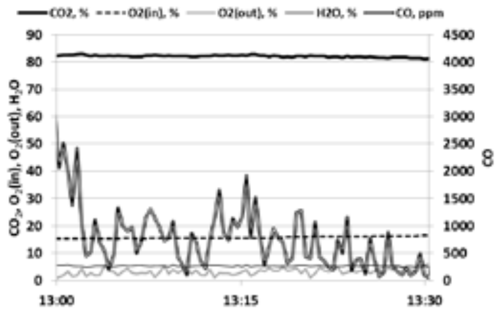

Fig. 1. Measured composition of flue gas during combustion of “Ziemowit” hard coal under pressure of 4 bar

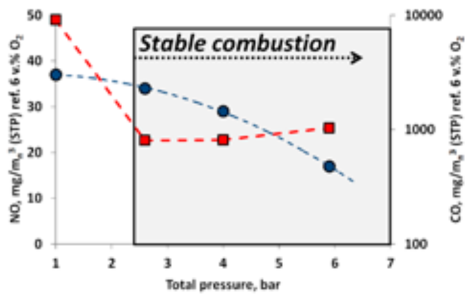

Fig. 2. The comparison of NO and CO emissions during combustion of “Ziemowit" hard coal (circles NO, squares CO)

Table 2. Average measured process parameters during combustion of “Ziemowit" hard coal

Press., bar	Press. drop, mbar**	$T_{freeboard}$, °C	O_2 (in), v.%	O_2 (out), v.%	CO, mg/m_n^3 (STP)	NO, mg/m_n^3 (STP)	N_2O, mg/m_n^3 (STP)	HCl, mg/m_n^3 (STP)	Fuel feed, kg/h	Velocity, m/s***	CH_4, ppm
1	16.9	720	16.4	2.7	9110*	37	121	291	0.47	1.4	111
2.6	13.8	820	16.3	3.6	800	34	89	278	0.42	0.6	7
4	14.0	883	15.5	3.1	809	29	23	216	0.79	0.5	23
5.9	18.6	910	14.8	2.1	1030	17	18	265	0.92	0.5	7

* Average value at high deviation of 2870 mg/m_n^3

** Pressure drop measured as the difference between the pressure under the sieve (gas distributor) and the end of the reactor

*** velocity calculated for conditions (pressure and temperature) in the bed

Ash issues

When combustion at low oxygen-concentration inlet is considered (higher probability of local reducing atmosphere), ash-related issues should be taken into account. It is known that ash fusions temperatures under reducing conditions are generally lower than those observed under oxidizing conditions (Wang and

Massoudi 2013). The composition of ash is an important parameter influencing the slagging and fouling risk. These undesired processes cause such negative effects like ash deposition on heat exchange surface and high temperature corrosion (Ściążko, Zuwała et al. 2006). Pronobis (Pronobis 2005) reviewed indexes that can be useful to determine the risk of slagging and fouling. Some indexes were also tested by Teixeira et al (Teixeira, Lopes et al. 2012). The characteristic values of the indexes are presented in Table 3. Ash composition, fusibility (oxidative and reducing atmosphere) and characteristic indexes to determine the risk of slagging and fouling for Ziemowit hard coal are presented in Table 4. Ash fusion characteristic (AFC), ash fusion temperature (AFT) depends on chemical and mineral composition. It was suggested by Vassilev et al. (Vassilev, Baxter et al. 2013) that temperature of hemisphere HT can be classified into five groups, i.e. (1) very low (<1000 °C), (2) low (1000–1200 °C), (3) moderate (1200–1400 °C), (4) high (1400–1600 °C), (5) and very high (>1600 °C). Ash from Ziemowit hard coal is classified into "moderate" group. However, the values are close to "high" range. Some indexes riched low or medium values. However, fouling index F_u, chloride content and ash fusibility index, AFI present high risk probability of fouling and slagging. Undoubtedly, the negative parameter of coal is chlorine content that riches quite high value (i.e. 0.39 % at "as received" state). Chloride compounds contribute in negative phenomena inside boiler i.e. alkali-induced slagging (by condensation of alkali chlorides), agglomeration, chloride-associated corrosion (by the impact of Cl_2 and alkali chlorides in gaseous, solid/deposited and liquid/molten phase) (Niu, Tan et al. 2016). One of the possible way to inhibit the negative effect of chlorine impact is sulphur addition to a combustion chamber (Silvennoinen and Hedman 2013). It was noticed that corrosion risk is not high when sluphur/chloride ratio (S^r/Cl^r) is higher than 2.0 (Ściążko, Zuwała et al. 2006). In case of Ziemowit hard coal S^r/Cl^r ratio was 3.04. However when sulphur retention in ash is taken into account (i.e. combustible sulphur is used in the S^r/Cl^r ratio) the values of S^r/Cl^r is 2.41.

Table 3. The characteristic values of indexes to determine the risk of slagging and fouling, according to Pronobis (Pronobis 2005) and Teixeira et al (Teixeira, Lopes et al. 2012)

Parameter	Value or range	Evaluate
$R_{(b/a)}$	<0.75	higher HT and FT, inhibited danger of slagging
	≈0.75	the lowest level of HT and FT, the strongest slagging probability
	0.75-2.0	HT and FT grow with increasing values of $R_{(b/a)}$
R_S	<0.6	low slagging inclination
	0.6-2.0	medium slagging inclination
	2.0-2.6	high slagging inclination
	>2.6	extremely high
Fouling index F_u	<0.6	low fouling inclination
	0.6-40	high fouling inclination
	>40	extremely high, tendency of sintering of deposits
S_R ratio-slag viscosity	> 72	low slagging inclination
	65-72	medium slagging inclination
	<65	high slagging inclination
Chloride content (as received state, Cl^r)	<0.2	low slagging inclination
	0.2-0.3	medium slagging inclination
	0.3-0.5	high slagging inclination
	>0.5	extremely high slagging inclination
Ash fusibility index, AFI	<1149°C	severe slagging potential
	1149-1232°C	high slagging potential
	1232-1343°C	medium slagging potential
	>1343°C	low

Table 4. Ash composition, fusibility (oxidative and reducing atmosphere) and characteristic indexes to determine the risk of slagging and fouling for Ziemowit hard coal

Fuel		HC
SiO_2	%	54.87
Al_2O_3	%	24.22
Fe_2O_3	%	7.42
CaO	%	2.9
MgO	%	2.33
P_4O_{10}	%	0.16
SO_3	%	2.49
Mn_3O_4	%	0.12
TiO_2	%	1.02
BaO	%	0.08
SrO	%	0.02
Na_2O	%	1.96
K_2O	%	2.41
sum	%	100
Temperature of (°C)	reducing atmosphere	oxidative atmosphere
initial deformation (IDT)	880	920
softening (ST)	1270	1270
hemisphere (HT)	1350	1380
fluid (FT)	1390	1420
AFI		
value	974	1012
evaluation	severe slagging potential	severe slagging potential
other ash indexes		
R(b/a)	value	0.16
	evaluation	inhibited danger of slagging
Rs	value	0.29
	evaluation	low slagging inclination
Fu	value	0.94
	evaluation	high fouling inclination
Sr	value	81.3
	evaluation	low slagging inclination
Cl r	value	0.390
	evaluation	high slagging inclination

CONCLUSIONS

The oxy-fuel combustion of Ziemowit hard coal under higher pressure was analyzed. The benefits of lower emission (i.e. NO and CO) and stabilization of combustion were observed under higher pressure (in range of 2.5-6 bar). Ultra-low NO emission was obtained (i.e. below 20 mg/m_n^3 (STP) calculated for NO and below 31 mg/m_n^3 (STP) calculated for NO_2). When combustion at low oxygen-concentration inlet is considered, ash-related issues should be taken into account. Slagging and fouling indexes were used to determine the risk of these negative effects. Some indexes indicates low probability of “ash-problems”. However, some indexes, i.e. fouling index F_u, chloride content and ash fusibility index, AFI present high risk probability of fouling and slagging. Instead of benefits related to ultra-low emission, ash issues should be considered during scaling up of oxy-fuel technology in future.

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