

# **Effect of Coal Properties and Operation Conditions on Flow Behavior of Coal Slag in Entrained Flow Gasifiers: A Brief Review**

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## EXECUTIVE SUMMARY

Integrated gasification combined cycle (IGCC) is a potentially promising clean technology with an inherent advantage of low emissions, since the process removes contaminants before combustion instead of from flue gas after combustion, as in a conventional coal steam plant. In addition, IGCC has potential for cost-effective carbon dioxide capture. Availability and high capital costs are the main challenges to making IGCC technology more competitive and fully commercial. Experiences from demonstrated IGCC plants show that, in the gasification system, low availability is largely due to slag buildup in the gasifier and fouling in the syngas cooler downstream of the gasification system.

In the entrained flow gasifiers used in IGCC plants, the majority of mineral matter transforms to liquid slag on the wall of the gasifier and flows out the bottom. However, a small fraction of the mineral matter (as fly ash) is entrained with the raw syngas out of the gasifier to downstream processing. This molten/sticky fly ash could cause fouling of the syngas cooler. Therefore, it is preferable to minimize the quantity of fly ash and maximize slag. In addition, the hot raw syngas is cooled to convert any entrained molten fly slag to hardened solid fly ash prior to entering the syngas cooler. To improve gasification availability through better design and operation of the gasification process, better understanding of slag behavior and characteristics of the slagging process are needed.

Slagging behavior is affected by char/ash properties, gas compositions in the gasifier, the gasifier wall structure, fluid dynamics, and plant operating conditions (mainly temperature and oxygen/carbon ratio). The viscosity of the slag is used to characterize the behavior of the slag flow and is the dominating factor to determine the probability that ash particles will stick. Slag viscosity strongly depends on the temperature and chemical composition of the slag. Because coal has varying ash content and composition, different operating conditions are required to maintain the slag flow and limit problems downstream.

This report briefly introduces the IGCC process, the gasification process, and the main types and operating conditions of entrained flow gasifiers used in IGCC plants. This report also discusses the effects of coal ash and slag properties on slag flow and its qualities required for the entrained flow gasifier. Finally this report will identify the key operating conditions affecting slag flow behaviors, including temperature, oxygen/coal ratio, and flux agents.

## 1. INTRODUCTION

Coal is an abundant energy resource in the United States, generating 45% of domestic electricity in 2009 (EIA 2011). However, burning coal also carries environmental concerns, such as sulfur compounds, and future concerns regarding carbon dioxide. Clean coal technologies are needed to provide better environmental performance at a low cost, enabling power plants to continue using coal for electricity generation. Integrated gasification combined cycle (IGCC) is a promising clean technology with an inherent advantage of low emissions, since the process removes contaminants from fuel gas with less volume and high partial pressure of contaminants rather than removing contaminants from flue gas after combustion, as in conventional coal plants (Maurstad 2005). In addition, IGCC has potential for cost-effective carbon dioxide capture (Falcke et al 2011).

In an IGCC system, coal is gasified to synthesis gas (syngas), which consists mainly of hydrogen and carbon monoxide. Syngas is then converted to electricity using a combined cycle that includes a gas turbine, a heat recovery steam generator (HRSG), and a steam turbine. Coal gasification has been commercially used to produce liquid fuels (Sasol in South Africa for 50 years [Sasol Synfuels International 2005]) and chemicals (Eastern Chemicals in the United States for 20 years [Trapp et al 2004]). The combined cycle technology is similar to those used in commercial modern natural gas (methane) fired power plants. In the United States, two commercial/demonstration IGCC power plants—Tampa Electric’s 250 MW Polk station and Duke Energy’s 262 MW Wabash River Repowering project—were built in the 1990s, partially through federal funds (DOE 2004). For IGCC technology to become more competitive and fully commercial, availability (defined as plant operation time during a certain period of time, usually 1 year) and high capital costs are the main challenges (Maurstad 2005, Trapp 2004).

Low availability of the IGCC plant is due to outages, planned and unplanned, of four major sections of combined cycle power units (CCU): gasification, air separation unit (ASU), and acid gas removal and sulfur recovery (AGR-SRV)/gas treatment (Higman et al 2006). Between 2001 and 2003, the cause of unplanned outages were approximately 17% CCU, 6% gasification, 3% ASU, and 2% gas treatment, based on the experience of four coal-based IGCC plants: Tampa Electric’s Polk Power Station (United States), Wabash River (United States), NUON (Netherlands), and ELCOGAS (Spain) (Higman et al 2006, EPRI 2007). In the gasification section, the major cause of unplanned downtime is fouling and plugging in the syngas cooler system and, significantly, in the slurry feed of IGCC plants (Higman et al 2006, EPRI 2006, TEC 2002). High amounts of unconverted carbon from the gasifier and failure of the refractory lining are issues in entrained flow gasifiers (TEC 2002).

In entrained flow gasifiers used in IGCC applications, the majority of the ash is melted and deposited on the wall of the gasifier, forming a liquid slag, which flows out the bottom of the gasifier and finally solidifies in a water bath. However, a small fraction of the ash is entrained as fly ash with the raw syngas out of the gasifier to downstream processing (Maurstad 2005). Molten/sticky ash in the fly ash could cause fouling of the syngas cooler (Brooker 1993, Brooker and Oh 1995, Erickson et al 1995, Maurstad 2005). The sticky ash particles deposit on the heat transfer surfaces of the syngas cooler and adhere to each other as well as to non-sticky ash particles. Therefore, it is preferable to minimize the quantity of fly ash and maximize slag. In addition, the hot raw syngas is

cooled to convert any entrained molten fly slag to hardened solid fly ash prior to entering the syngas cooler (Maurstad 2005). To improve gasification availability by improving the design and operation of the gasification process, a better understanding of slag behavior and the characteristics of the slagging process through both experiments and computer modeling are needed.

Slagging behavior is affected by the properties of both gaseous and solid phases, gasifier wall structure, fluid dynamics, and operating conditions (such as temperature, oxygen/carbon ratio, steam/oxygen ratio, pressure, and residence time). The deposition of ash particles to the wall is a function of carbon conversion that is affected by operating conditions (Li et al 2010). In general, the slag rheology, like any fluid, is a function of its molecular structure and dynamics (Duchesne et al 2010). The viscosity of the slag is used to characterize slag flow behaviors and is the dominating factor that determines the ash particle sticking probability, among many factors, such as particle velocity, surface tension, temperature, size, and impact angle (Wang and Harb 1997). Under reducing conditions (gasification), the viscosity at a given temperature is generally lower than under oxidizing atmospheres (conventional combustion). Slag viscosity strongly depends on temperature. At elevated temperature, slag acts as a Newtonian fluid and its viscosity usually decreases logarithmically as temperatures increase. At lower temperatures, crystallization or the separation of immiscible liquids may cause a dramatic increase in viscosity and non-Newtonian behaviors.

To ensure continuous slag flow, a slag viscosity less than 25 Pa·s (250 poise) is necessary (Higman and van der Burgt 2008). The operating temperature of the gasifier is strongly dependant on slag viscosity. To reduce slag viscosity, one option is to raise the gasifier operating temperature, which requires more oxygen and burns more fuel, thereby increasing cost and lowering process efficiency. Another option is to add a flux or blend coals with low fusibility since viscosity is affected by the chemical compositions of ash (Patterson and Hurst 2000, Duchesne et al 2010). Flux usage and its cost are proportional to ash content (Higman and van der Burgt 2008). In addition, all the ash must be melted, and more heat is required for greater ash content, thereby reducing cold gas efficiency (Higman and van der Burgt 2008).

Slag flow is controlled by the viscosity of the slag, which in turn depends on the slag temperature and slag chemical composition. Changes in the coal result in changes in the quantity and composition of ash and, consequently, cause changes in the flow of the slag within and exiting the gasifier, as well as changes in downstream processing. Therefore, the effect of coal properties, such as ash content, ash composition, and operating conditions related to coal conversion on slag flow behavior (such as slag viscosity) should be investigated to improve the availability of IGCC plants. The objectives of this report are to briefly introduce type and operating conditions of entrained flow gasifiers used in IGCC plants; discuss the effects of ash properties, coal conversion, and flux on slag flow behaviors; characterize ash viscosity as it relates to temperature and ash composition through experiments and empirical models; and suggest the coal quality criteria and operating conditions for use in entrained flow slagging gasifiers.

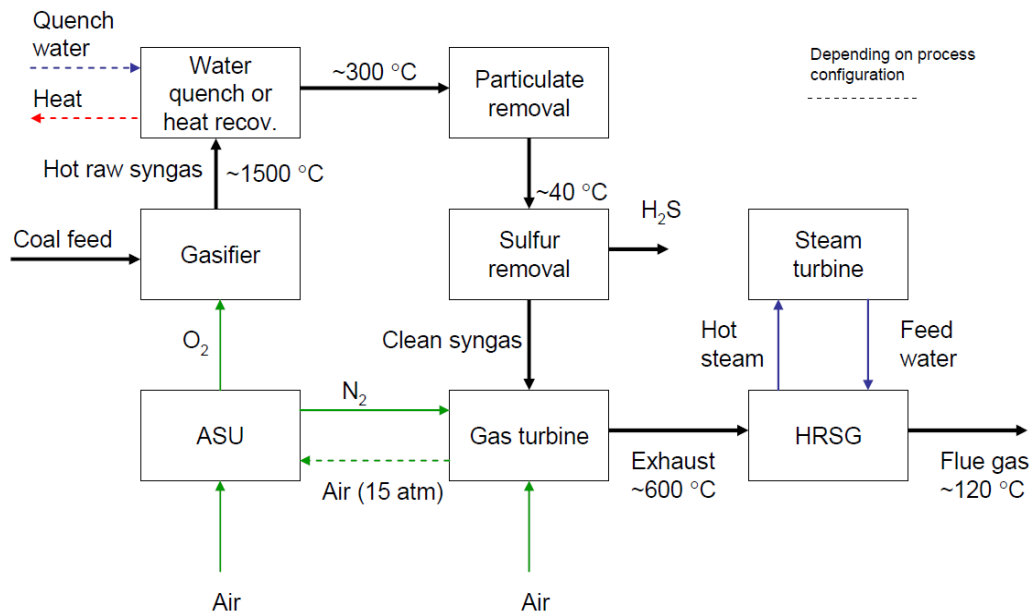
## 2. FUNDAMENTALS OF COAL GASIFICATION IN IGCC

### 2.1. IGCC Process Description

The IGCC process typically consists of a gasification system and a power system. The gasification system includes coal preparation, a gasification reactor (gasifier), an ASU, gas cooling, and gas clean up. A water-gas-shift reactor and carbon dioxide separation unit may be added to obtain hydrogen-rich syngas and carbon dioxide for sequestration. Three major components of the power system are a combustion turbine (CT; also called a gas turbine, GT), a HRSG, and a steam turbine. The power system is similar to those used in commercial modern natural gas (methane) fired power plants.

In recently demonstrated IGCC plants (without carbon dioxide capture), coal is pulverized and fed to a gasifier one of two ways: dry fed (mixed with nitrogen) or wet fed (mixed with water) (Figure 2) (Maurstad 2005). Fluxing agents or additives, such as lime, may be added to optimize the ash melting point and adjust flow characteristics. Gasifying agents of steam and a limited amount of oxygen (from the ASU) flow to the gasifier. Under reducing conditions, the coal is gasified into hot raw syngas at a high temperature (around 1500 °C) and high pressure in gasifiers that have refractory walls or membrane walls. Ash is melted to a liquid slag and removed from the bottom of the gasifier, which, for the majority of IGCC project applications, is an entrained flow type.

The hot raw syngas is cooled to ~900 °C by quenching and further cooled by syngas cooler (sensible heat recovery) to ~300 °C for the gas cleaning up process to remove particulates and sulfur. Before leaving the gasifier, the quench solidifies the slag to avoid fly ash with sticky surfaces. The sensible heat contained in the hot raw syngas is recovered using a syngas cooler for the steam turbine. The clean syngas, which consists of hydrogen and carbon monoxide, is burned in a gas turbine. The hot exhaust



**Figure 1. IGCC process without CO<sub>2</sub> capture (Maurstad 2005).**

from the gas turbine passes through a HRSG to generate steam for the steam turbine. Electricity is generated from both the gas- and steam-turbine generators.

## 2. 2. Chemistry of Coal Gasification

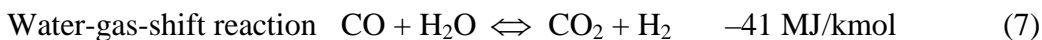
Gasification is a thermal chemical process in which carbonaceous materials (coal, petroleum coke, biomass, heavy oil, etc.) are converted to syngas by partial oxidation with air or oxygen, and/or steam. Exothermic oxidation reactions provide the necessary heat for the endothermic reactions. Steam or water is added to increase the amount of hydrogen in the product gas.

The coal gasification process mainly includes pyrolysis/devolatilization, combustion, and gasification. The devolatilization takes place at low temperature (350–800 °C) as coal particles are heated up and mainly produces gases, tars, and char (Higman and van der Burgt 2008). Light gases include hydrogen, carbon monoxide, carbon dioxide, methane, acetylene, ethylene, and so forth (Smoot and Smith 1985). Tars are organic components with molecular weights of 200–2000 and liquefy at room temperature (Cortés et al 2009). Char consists of non-reacted carbon and ashes. Gasification takes place at temperatures of 800–1800 °C (Higman and van der Burgt 2008). During gasification, the volatiles (gases and tars) and char react with the oxidants. The partial combustion of solid carbon and volatiles generate heat for the gasification reactions. However, volatile combustion may not complete in the gasification environment if oxygen is limited. In entrained flow gasifiers, devolatilization and gasification take place simultaneously. The principle gasification reactions are as follows (Higman and van der Burgt 2008):

Reactions in the solid phase:



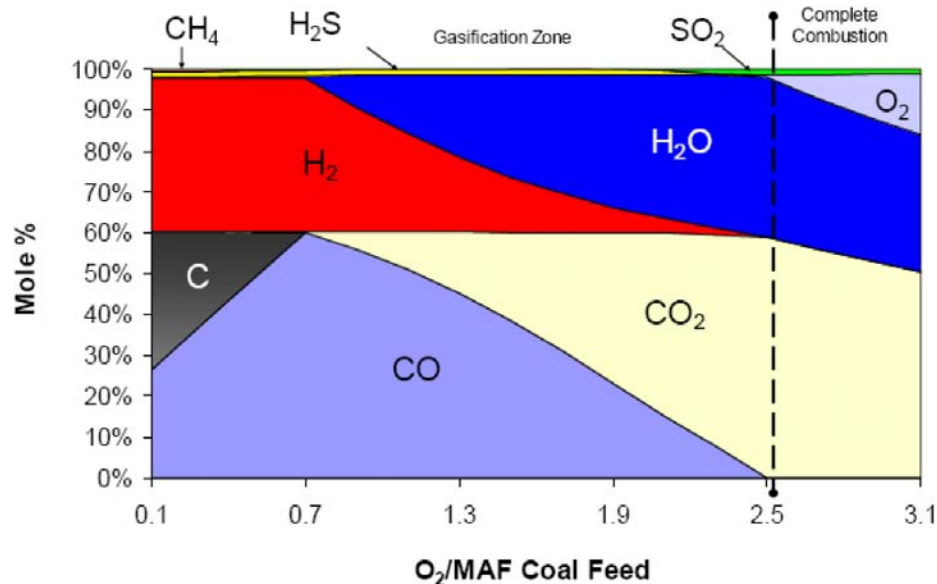
Reactions in the gas phase:



The water gas reaction (2) is a main gasification reaction. Most gasification processes rely on a balance between solid carbon partial oxidation (1) and the water gas reaction (2) (Higman and van der Burgt 2008). The Boudouard reaction may be inhibited by the carbon monoxide produced during partial oxidation (1) and the water gas reaction (2). The methanation reaction is normally very slow and at low temperature unless it occurs under high pressure or in the presence of a catalyst. The water-gas-shift reaction

occurs at low temperature in the presence of a catalyst and is used to adjust the hydrogen/carbon monoxide ratio in the gas.

Compositions of gases produced from gasification are significantly controlled by the oxygen/coal ratio in the gasifier. Figure 2 shows the products of the reaction as a function of the oxygen/coal ratio from gasification conditions to full combustion for Illinois #6 coal by dry feeding (NETL 2011).



**Figure 2.** The products of reactions as a function of oxygen/coal ratio from gasification conditions to full combustion of Illinois #6 coal by dry feeding (NETL 2011).

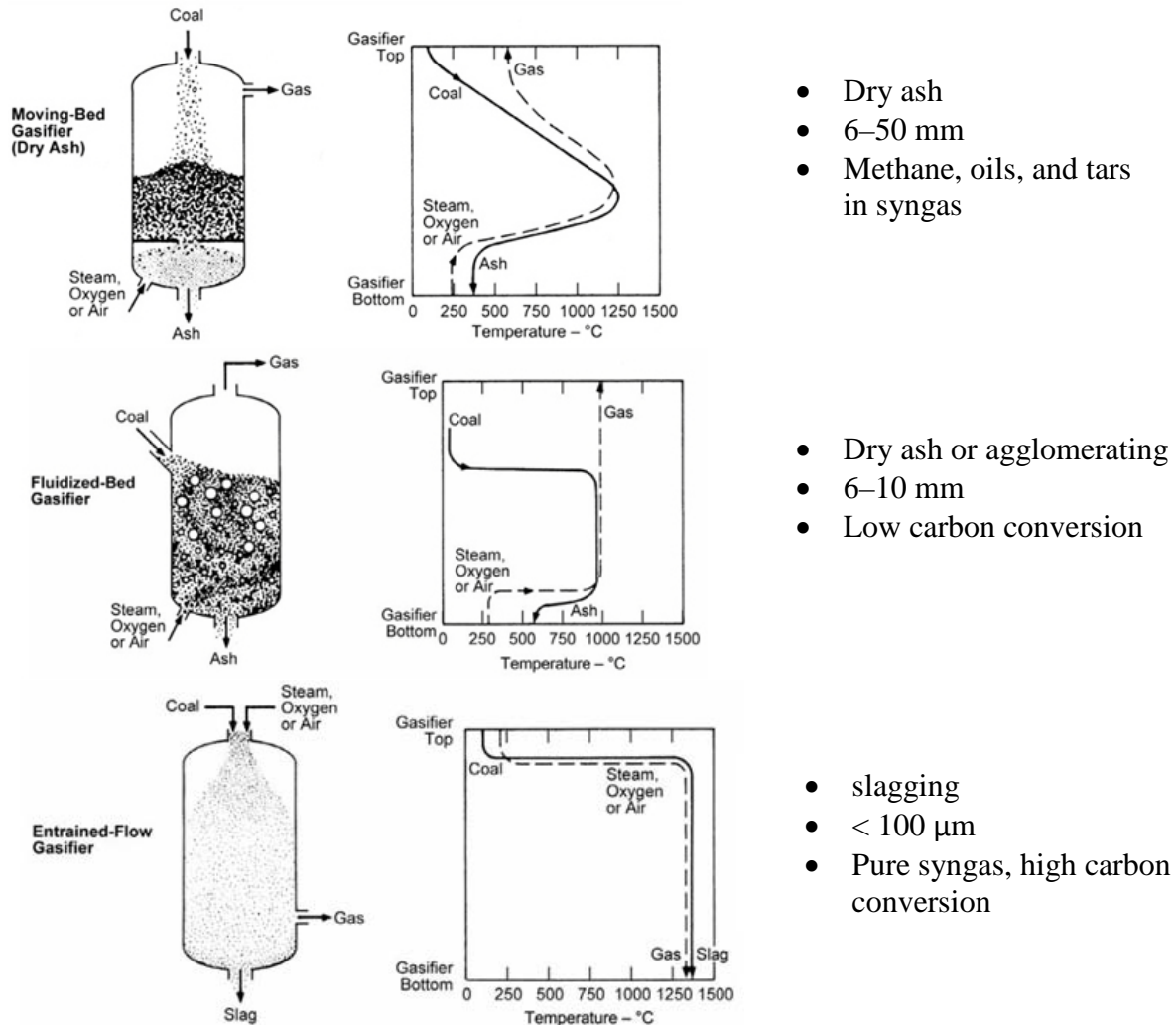
## 2. 3. Gasifiers

### 2. 3.1. Classification of Gasifiers

Many types of gasifier have been developed. Based on flow conditions in the gasifier, three categories of gasifiers are fixed/moving bed, fluidized bed, and entrained flow (slagging) gasifiers. The flow conditions of coal, gasifying agents (steam and oxygen), and syngas are illustrated in Figure 3 (NETL 2011). Figure 3 also lists important characteristics (Higman and van der Burgt 2008).

The four major commercial gasification technologies, in order of decreasing capacity, are Sasol-Lurgi dry ash, GE (originally developed by Texaco), Shell, and ConocoPhillips E-gas (originally developed by Destec) (Maurstad 2005). The Sasol-Lurgi gasifier is a fixed/moving bed type gasifier with dry ash and has been used extensively to produce synfuel by Sasol in South-Africa. GE, Shell, and E-gas gasifiers are entrained flow type and are used in the four major commercial-scale coal-based IGCC demonstration plants of Tampa Electric's Polk Station (Florida, United States), Wabash River (Indiana, United States), NUON(formerly Demkolec; Buggenum, The Netherlands), and ELCOGAS (Puertollano, Spain) (EPRI 2006).





**Figure 3. Classification and characteristics of the three major types of gasifier (NETL 2011).**

Fluidized-bed gasifiers are less developed compared to the two other types of gasifiers. KBR's transport integrated gasification (TRIG) technology is a fluidized-bed type gasifier. The Kemper County IGCC project will use TRIG that is potentially suited for low rank coals with high moisture and ash contents (Maurstad 2005). The project has been under development for the past decade by Southern Company, KBR Inc., and the U.S. Department of Energy.

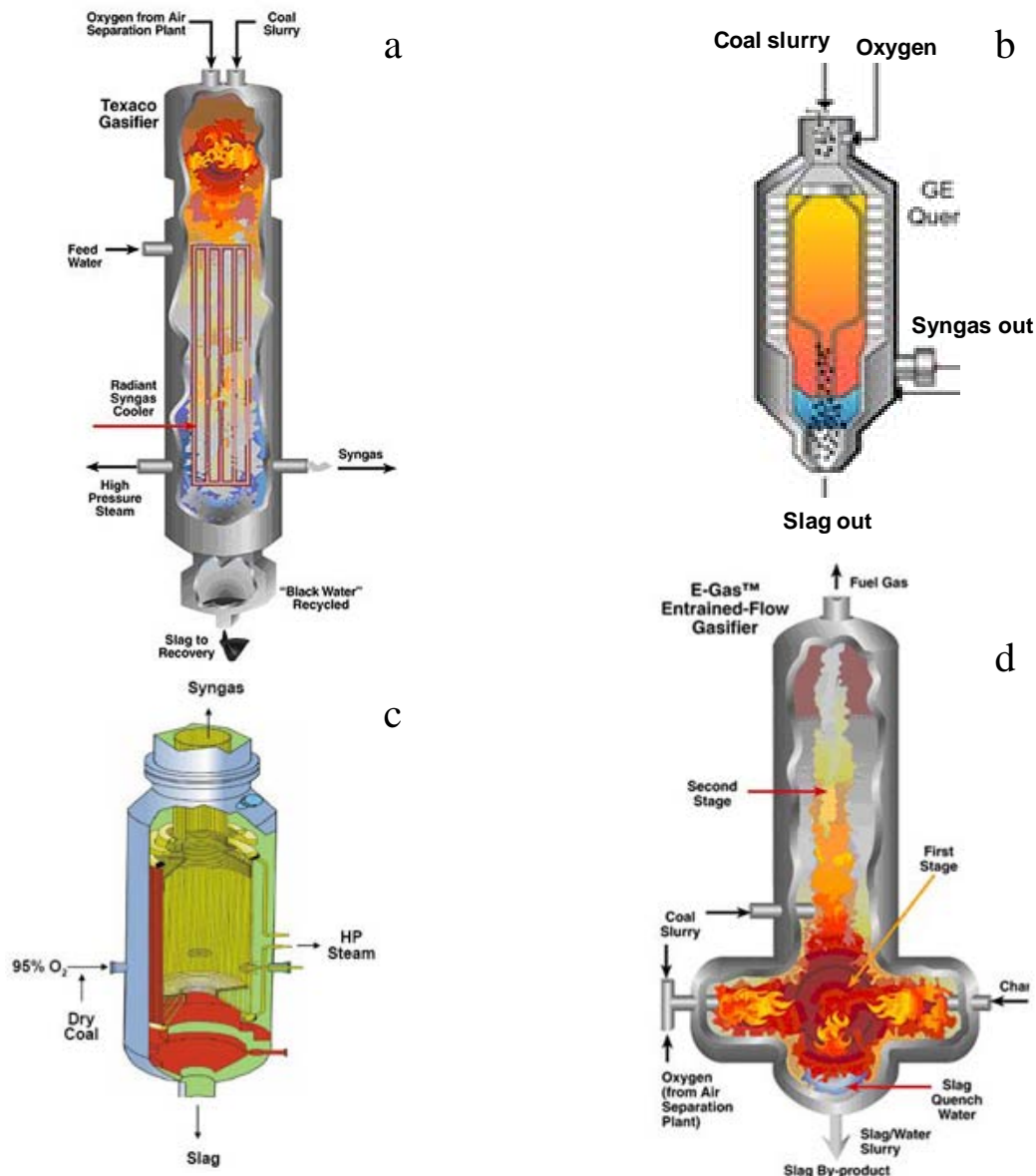
### 2.3.2. Characteristics of Major Commercial Entrained Flow Technologies

GE Energy, Shell, and E-gas gasifiers are the major commercial entrained flow technologies used to generate raw syngas (Table 1) (Maurstad 2005). The raw syngas generation process consists of hot raw syngas generation, quenching, and syngas cooling. In the hot raw syngas generation process, coal is fed to a gasifier by dry feed (Shell at the bottom of the gasifier) or slurry feeding (GE at the top of the gasifier, and E-gas at the bottom of the gasifier) (Figure 4) in either a single stage (Shell and GE) or two stages (E-gas) (NETL 2011). Gasifying agents are supplied via the top of the gasifier (Shell,

oxygen and steam; E-gas, oxygen) or the bottom of the gasifier (GE, oxygen). The hot raw syngas is generated at high temperature (around 1500 °C) and pressure in the gasifiers that have refractory wall (GE and E-gas) or membrane wall (Shell). Ash is formed and melted, the majority of which deposits on the wall of the gasifier and leaves the gasifier in a liquid flow as slag. The remaining ash/fly ash in liquid form is entrained in the syngas and may create a potential fouling problem for downstream process equipment, such as the syngas cooler. Therefore, prior to the hot syngas leaving the gasifier, it is quenched to solidify the melting fly ash and avoid the fly ash with sticky surfaces. The quenched syngas leaves the gasifier at its top (Shell and

**Table 1. Characteristics of major commercial entrained flow technologies (Maurstad 2005, and EPRI 2007).**

| Gasification technology/<br>Design feature | GE Energy<br>with radiant<br>cooling                           | GE Energy<br>with water<br>quench                              | Shell  | E-gas  |
|--|--|--|--|--|
| Feed system                                | 60 to 70% coal/water slurry; rod mills; slurry pumps; top feed | 60 to 70% coal/water slurry; rod mills; slurry pumps; top feed | Dry coal (~2% MC)/N <sub>2</sub> ; roller mills; lock hoppers; bottom feed | 60 to 70% coal/water slurry; rod mills; slurry pumps; bottom feed: 1st stage 80% and 2nd stage 20% |
| Stage and flow                             | Single stage downflow  | Single stage downflow  | Single stage upflow  | Two stage upflow   |
| Wall                                       | Refractory   | Refractory   | Membrane   | Refractory   |
| Pressure (bar)                             | 30–80  | 30–80  | 30–40  | ~27  |
| Hot syngas temperature (°C)                | 1260–1500  | 1260–1500  | 1360–1650  | 1st stage 1340–1400; 2nd stage 1038  |
| Syngas quenching method                    | Radiant cooling then fire tube syngas cooler                   | Water quench (no syngas cooler)                                | Recirculated cold syngas quench then water tube syngas cooler              | Chemical quench then fire tube syngas cooler   |
| Quenched syngas temperature (°C)           | ~800   | ~300   | ~900   | ~1038  |



**Figure 4. Characteristics of the major commercial entrained flow gasifiers: a. GE gasifier with radiant cooling, b. GE gasifier with water quench, c. Shell gasifier, and d. E-gas gasifier (NETL 2011).**

E-gas) or its bottom (GE). The sensible heat of the quenched syngas with a temperature around 900 °C (a, c and d in Figure 4) is recovered in a syngas cooler to produce steam for the steam turbine except the case where syngas is quenched using water to around 300 °C (b in Figure 4).

There are four major syngas quenching methods: radiant syngas cooling (GE), water quench (GE), gas recycle quench (Shell), and chemical quench (E-gas) (Maurstad 2005). In radiant syngas cooling, the hot gas flows into a radiant boiler where steam is generated for a turbine. Slag is dropped into a water bath at the bottom of the gasifier. The boiler is expensive and may have fouling problems. In the water quench, the hot gas and slag are cooled. The sensible heat of the syngas is recovered to vaporize water, and

the syngas is saturated with water vapor. In the gas recycle quench, cooler syngas with a temperature around 300 °C from downstream is recycled to the gasifier outlet to cool the syngas from 1500 °C to 900 °C by mixing hot and cold syngas. In the chemical quench, the hot syngas that is generated in the first stage of gasification is quenched by the second stage of gasification. From the first stage, the sensible heat of the hot syngas, with a temperature around 1400 °C, is used to gasify the coal at the second stage of gasification. The second gasification is a non-slag process at lower temperature and may make downstream gas clean up for tar more complex. In addition, Ni et al (2011) simulated and compared all types of syngas cooling systems. Numerical models were developed to predict the multiphase flow and temperature of the quenching gas and the wall of the gasifier.

Two types of syngas coolers are fire tube boilers (GE and E-gas) and water tube boilers (Shell) (Maurstad 2005, EPRI 2007). In the fire tube boiler, which costs less than the water tube boiler, hot syngas flows inside of the tubes and steam is generated on the outside. The water tube boiler works in the opposite way.

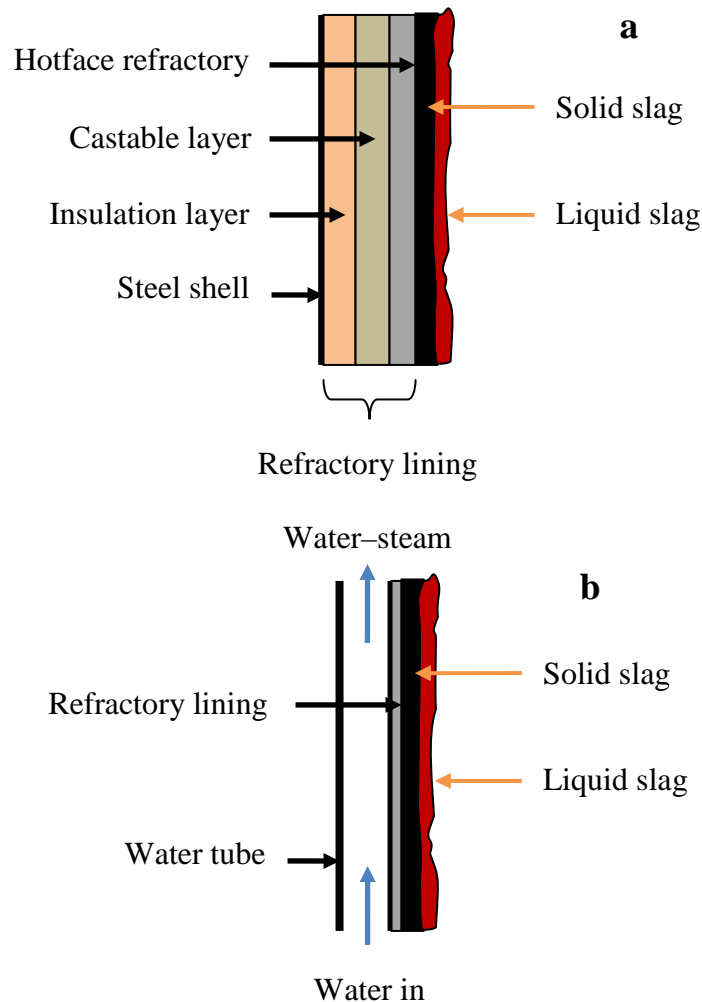
### **2.3.3. Gasifier Wall Fundamentals**

Two types of refractory linings and membrane walls are used in entrained flow slagging gasifiers to protect the steel walls and reduce heat loss (EPRI 2006). ConocoPhillips E-gas and GE Energy gasifiers use refractory linings, while membrane walls are used in the Shell gasifier. Refractory lining typically consists of three layers: an insulation layer of silica firebrick (200–300 °C), a layer of castable bubble alumina, and a hotface refractory (Figure 5a) (EPRI 2006, Higman and van der Burgh 2008). The inner hotface refractory is usually a chromium oxide-based and/or zirconium oxide-based brick (Higman and van der Burgh 2008). It is worn by chemical corrosion (such as silica in the ash) and erosion (hot liquid slag). Although the refractory lining itself is inexpensive, repairs and/or replacements are frequent and costly. For example, Tampa Electric's Polk Power Station has a 20–30 day planned outage every 2 years (TEC 2002).

The water-cooled membrane walls consist of high-pressure tubes, flat steel bridges that connect the tubes, and a thin layer of castable refractory (usually silicon carbide) (Figure 5b) (Higman and van der Burgh 2008, Cortés et al 2009). Solid slag covers the wall surface, providing a protective layer. Membrane walls have a complex and expensive cooling and control system, but the walls have an estimated lifetime of about 20 years (Higman and van der Burgh 2008). In addition, the heat loss with the membrane is 2–4% of the heating value of the coal and is higher than heat loss with refractory linings, which is less than 1%.

## **2. 4. Entrained Flow Gasifier Operating Conditions**

Entrained flow gasifiers with high temperature and high pressure have been selected for the majority of IGCC projects (Table 1). A major advantage of using high-temperature entrained flow gasifiers is to generate syngas that is free of oils and tars so the related problems can be avoided (Maurstad 2005). The high reaction rate at the high temperature also allows single gasifiers to be built with large gas outputs sufficient to fuel large commercial gas turbines. High carbon conversion and low methane production are other benefits of high temperature. High-pressure syngas can be directly fed to a gas



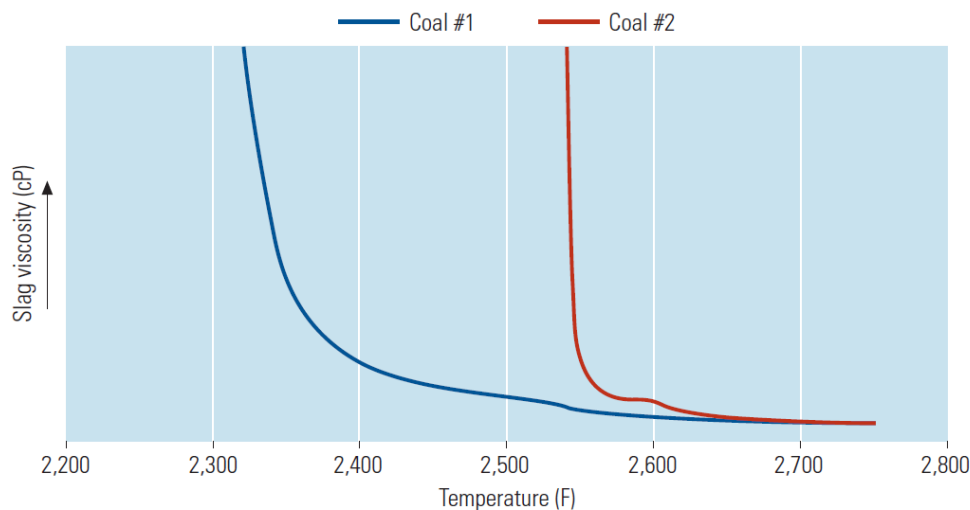
**Figure 5. a. Refractory lining (EPRI 2006) and b. Membrane wall (Cortés et al 2009).**

turbine to avoid large auxiliary power losses for syngas compression. It reduces capital cost since it increases the capacity of the gasifier in volume. In addition, downstream syngas cleaning processes, such as carbon dioxide capture, will be more efficient because of the increased partial pressure.

Gasifier operating conditions are determined by carbon conversion, ash slagging temperature or ash flow temperature of the coal, slag viscosity presented using  $T_{25}$  temperature (the temperature at which the slag viscosity is 25 Pa·s), and temperature of critical viscosity ( $T_{cv}$ ). High amounts of unconverted carbon in the ash affect ash deposition on the wall of gasifier (Li et al 2010). Fly ash with high unconverted carbon causes difficulties in downstream processing (EPRI 2006). Low temperature and high slag viscosity may solidify the slag and cause slag blockage (EPRI 2006). All these difficulties can be overcome by sending more oxygen to the gasifier to burn more coal and generate more heat. However, this reduces the overall efficiency of an IGCC system by increasing the parasitic power load of the air separation unit, reducing syngas yield

and, when a refractory-lined gasifier is used, shortening the life of the refractory. For most coals, the ash melting point and slag viscosity are more constraining than carbon conversion considerations; therefore, the operating temperature of the gasifier is selected based on the ash slagging temperature or ash flow temperature and slag viscosity (EPRI 2006)—characteristics that are important for selecting the appropriate operation temperature of the gasifier to avoid either overfiring or solidifying the slag within the gasifier.

Based on the gasification experiences of Eastman Chemical Company, slag viscosity should be considered along with the ash melting temperature because coals with the same ash fusion temperature have different slag viscosities and, therefore, behave differently in the slag gasifier (Trapp et al 2004). As shown in Figure 6, coal #1 could run at a much lower temperature compared to coal #2, according to Eastman Chemical Company experiences. The slag of coal #2 would be very viscous even at 2550 °F. The high-temperature slag would wear the gasifier's refractory and reduce total gasification efficiency. Therefore, slag viscosity measurement is important in the gasifier along with ash slagging temperature.



**Figure 6. Slag viscosity versus temperature for two different coals with the same ash fusion temperatures (Trapp et al 2004).**

In the gasification process, oxygen is used to gasify solid carbon to syngas (such as reaction 1) and generate heat to drive other gasification reactions (2 and 3) along with the combustion of volatiles. In an ideal gasifier, an amount of oxygen would be injected to react with carbon in the coal, based on reaction (1). However, in a real gasifier, additional oxygen is needed to oxidize some of the carbon monoxide to generate heat, as described in reaction (5). In addition, more oxygen is added to increase the operating temperature in the entrained flow gasifier above the slagging temperature or melting point,  $T_{25}$  and  $T_{cv}$ , of the ash by burning more carbon (EPRI 2006).

To ensure near complete conversion to syngas, gasification typically operates above the stoichiometric oxygen/coal ratio. Compared to combustion, the amount of oxygen used in gasification is far less, typically less than half.

### 3. COAL PROPERTIES AND QUALITIES FOR ENTRAINED FLOW SLAGGING GASIFIERS

#### 3.1. Ash Content and Coal Composition

Compared to other factors, such as price, heating value, sulfur content, and availability, coal ash content is the most important factor in selecting coal because coal ash content has the most impact on the performance of a slagging gasifier (Trapp et al 2004). High-ash coals are not preferred because all the ash must be melted, which requires more heat and, therefore, more coal must be burned. As a result, cold gas efficiency reduces as ash content in the coal increases (Higman and van der Burgt 2008). Furthermore, if additional flux is required, the flux and its cost are proportional to the ash contents. By the economic limit, the ash content of coal (% mass) for a slurry feeding gasifier is generally about 20% (consider evaporating water by heat) and about 40% for a dry feeding gasifier (Higman and van der Burgt 2008).

**Table 2. Summary of criteria for coal ash properties for entrained flow gasifiers (Higman and van der Burgt 2008, Patterson and Hurst 2000).**

| Coal ash properties | Criteria for entrained flow gasifier  | Reference                     |
|---------------------|---|-------------------------------|
| Ash contents        | 20% slurry feeding, 40% dry feeding   | Higman and van der Burgt 2008 |
| Ash FT              | < 1400 °C is optimal but 1500 °C is acceptable; >1500 °C flux is needed   | Patterson and Hurst 2000      |
| Ash composition     | silica ratio $100 \text{ SiO}_2/(\text{SiO}_2 + \text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO}) < 80$ ; $\text{SiO}_2/\text{Al}_2\text{O}_3$ ratio of 1.6–2.0 is optimal for minimum flux amount | Patterson and Hurst 2000      |
| $T_{cv}$            | < 1400 °C is preferred  | Patterson and Hurst 2000      |
| Viscosity           | 5–25 Pa·s at 1400 to 1500 °C; Optimum 15 Pa·s   | Patterson and Hurst 2000      |

Ash content analysis is included in proximate analysis, which is basic coal chemical analysis along with ultimate analysis to characterize coal behavior for gasification and combustion (Erickson et al 1995). The proximate analysis of coal determines the distribution of products obtained when coal is heated under specified conditions. The products are separated into four groups: moisture, volatile matter generated during pyrolysis, fixed carbon (nonvolatile), and ash. Fixed carbon is determined by difference and does not represent pure carbon in the coal. Coal is a heterogeneous material and consists of organic (macerals) and inorganic (mineral matter) components (Ward 2002). The organic matter is primarily composed of carbon, hydrogen, and oxygen, with lesser amounts of nitrogen and sulfur that are related to environmental pollutants (Miller 2005). All these elements are determined by the ultimate analysis. The inorganic fraction is commonly identified as ash, subsequent to combustion and gasification, and causes problems (such as fouling) during coal

combustion and gasification. Ash consists of inorganic residues remaining after combustion at 700–750 °C (ASTM-3174). Ash content in proximate analysis indicates the amount of inorganic matter in situ in the coal structure and out-of-seam inorganic contamination. It is different with mineral matter composed of the unaltered inorganic continuants in coal (Speight 2005).

There are three forms of inorganic continuants distributed in coal: organically associated elements (other inorganic constituents), included (inherent) minerals that are closely associated with coal, and excluded mineral matter that readily separates from coal (Erickson et al 2004). The main minerals in coal include kaolinite, quartz, aluminosilicates, pyrite, dolomite, and calcite, along with unknown phases (Van Dyk 2009a). In the gasifier, mineral matter/inorganic components are transformed into vapors (such as vaporized inorganic components), liquids (melting ashes), and solids (Reid 1981, Erickson et al 2004, Matsuoka et al 2006, Van Dyk 2009a). The intermediate ash species, produced during gasification, deposit and form slag or become entrained ash and flow out of the gasifier with flue gas as fly ash (Erickson et al 2004); the slag and fly ash have different chemical compositions (Aineto et al 2006). The fly ash and volatile compounds flow out of the gasifier. The vaporized inorganic components may condense or the melting fly ash (not solidified by syngas quenching) may deposit in syngas coolers and cause different types of fouling (Ward 2002). In liquid phase slag, the major bonding components are derived from included minerals, such as calcium, in carbon-rich particles (Van Dyk 2009a). The ash formed during gasification is in the formation of chemically reduced species, such as sulfides and metals, which have different properties from their oxidized counterparts in combustion (Erickson et al 2004, Font et al 2005).

Chlorine content in coal is mostly less than 1% (wt) but may be as high as 2.5% (Higman and van der Burgt 2008). Chlorides' melting points are 350–800 °C and may deposit in the syngas cooler and foul the exchanger surface. Phosphorus is not a problem for coal gasification but it may cause a fouling problem when adding biomass with high phosphorus content in the feedstock (Higman and van der Burgt 2008).

### 3.2. Chemical Composition of Ash in Coal

The chemical composition of coal ash is an important factor in slagging gasifiers because it affects ash fusibility and slag viscosity. Silicon dioxide ( $\text{SiO}_2$ ), aluminium oxide ( $\text{Al}_2\text{O}_3$ ), ferric oxide ( $\text{Fe}_2\text{O}_3$ ), titanium oxide ( $\text{TiO}_2$ ), phosphorus pentoxide ( $\text{P}_2\text{O}_5$ ), calcium oxide ( $\text{CaO}$ ), magnesium oxide ( $\text{MgO}$ ), sodium oxide ( $\text{Na}_2\text{O}$ ), potassium oxide ( $\text{K}_2\text{O}$ ), and sulphur trioxide ( $\text{SO}_3$ ) are the major components of coal ash (Erickson et al 1995, Higman and van der Burgt 2008, Van Dyk 2009a). Ash compositions in mass percent are 50.1  $\text{SiO}_2$ , 23.3  $\text{Al}_2\text{O}_3$ , 6.4  $\text{Fe}_2\text{O}_3$ , 1.0  $\text{TiO}_2$ , 0.7  $\text{P}_2\text{O}_5$ , 8.1  $\text{CaO}$ , 2.7  $\text{MgO}$ , 0.4  $\text{Na}_2\text{O}$ , 0.8  $\text{K}_2\text{O}$ , and 6.1  $\text{SO}_3$  (Van Dyk 2009a). These components mainly contribute to the melting characteristics of the ash. All these components, specifically the calcium and iron contents are believed to be indicators for ash fusion properties. Calcium oxide in particular is an important factor in the viscosity properties of a slag (Van Dyk 2009b). As the calcium oxide content increases, the viscosity of slag increases. Trace components, such as mercury, chlorine, fluorine, etc., have a major effect on the environmental issues associated with coal usage.

The chemical composition of the ash can be determined by many methods, including inductively coupled plasma (ICP) spectroscopy–atomic emission spectrometry,



x-ray fluorescence spectroscopy (XRF), x-ray power diffractometry (XRD), and computer-controlled scanning electron microscopy (CCSEM) (Ward 2002, Speight 2005, and Matsuoka et al 2006, Van Dyk 2009a). Ash composition could be used to assess slag viscosities, flow temperatures, and flux requirements (Patterson and Hurst 2000). In addition, indices (slagging and fouling) based on ash compositions could be used to guide operations and evaluate coal quality for gasification or to describe ash slagging or fouling behavior. There are many proposed indices, but they only have limited application because they are not based on physical principles. The base-acid ratio  $R$  as  $(\text{CaO} + \text{MgO} + \text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O})/(\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{TiO}_2)$  can be used for all ranks of coal (Su et al 2003). Coal with  $R$  in the range of 0.4–0.7 has low fusibility temperature and higher slagging potential (Miller 2005). One of slagging index is  $R_s$  as  $R \times S$ , where  $S$  is the wt % sulfur in the dry coal. Some fouling indices are  $R_F$  as  $R \times \text{Na}_2\text{O}$  and chlorine content of coal to predict fouling of convective heating surface. For blended coals, the fouling index  $\text{Na}_2\text{O}$  (g/GJ) is better than  $R_F$  (Su et al 2003). Other factors are silica/alumina ratio ( $\text{SiO}_2/\text{Al}_2\text{O}_3$ ), iron/calcium ratio ( $\text{Fe}_2\text{O}_3/\text{CaO}$ ), a dolomite percentage (DP)  $100(\text{CaO} + \text{MgO})/(\text{Fe}_2\text{O}_3 + \text{CaO} + \text{SiO}_2 + \text{MgO} + \text{Na}_2\text{O} + \text{K}_2\text{O})$ , and a silica percentage (SP)  $100 \text{SiO}_2/(\text{SiO}_2 + \text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO})$ . For blended coal,  $\text{Fe}_2\text{O}_3/\text{CaO}$  molar ratio correlates to slagging propensity (Su et al 2001). All these indices and factors are from coal combustion and may be applied to coal gasification.

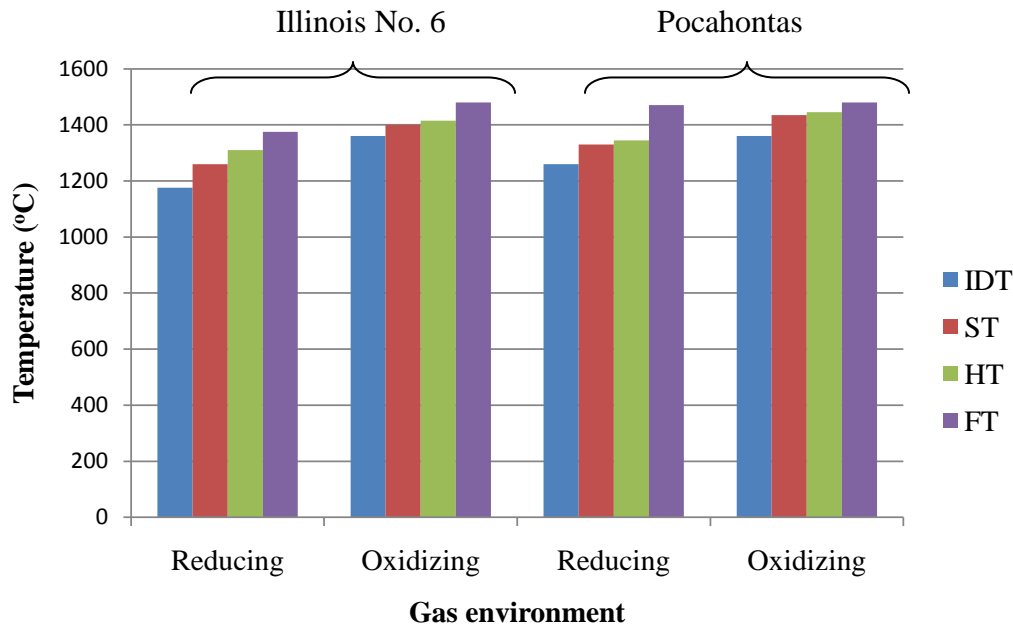
Patterson and Hurst (2000) studied an extensive range of Australian bituminous coals using laboratory tests for the entrained flow slagging gasifier. They proposed that a silica ratio  $100 \text{SiO}_2/(\text{SiO}_2 + \text{Fe}_2\text{O}_3 + \text{CaO} + \text{MgO}) < 80$  is required for entrained flow gasifiers. Flux (such as limestone) is required for coal with a higher silica ratio. To minimize the flux amount, a  $\text{SiO}_2/\text{Al}_2\text{O}_3$  ratio of 1.6–2.0 is optimal. Low-temperature ash deposition behavior (fouling) of various coals under gasification conditions was studied using a drop tube furnace (Xu et al 2009), with fouling related to  $R_{ba}$  ratio of  $\%(\text{MgO} + \text{CaO} + \text{K}_2\text{O})/\%(\text{SiO}_2 + \text{Al}_2\text{O}_3)$ .  $R_{ba}$  in the bottom layer of fouling in the probe is higher in the top layer. Since  $\%(\text{MgO} + 0.719\text{CaO})$  in coal is high, the coal demonstrates a propensity towards fouling.

### 3.3. Ash Fusibility and Ash Flow Temperature

The ash fusibility test (AFT) (ASTM D-1857) is designed to simulate the behavior of coal ash when it is heated in either a reducing or an oxidizing atmosphere. The test is the most accepted method of assessing the propensity of coal ash to slag and gives an average flow property. It measures approximate temperatures at which the ash cone will sinter (i.e., the solid ash particles will weld together without melting), melt, and flow. Four temperatures are reported: the initial deformation temperature (IDT), softening temperature (ST), hemispherical temperature (HT), and flow temperature (FT) (Speight 2005). For blended coal ash, thermodynamic analysis (TMA) was used to characterize ash fusibility because TMA temperatures changed with blend proportions of coals while AFT did not (Byrant et al 2000).

For gasification applications, the fusion characteristics of the ash should be determined under reducing conditions since these results are generally lower than those observed under oxidizing conditions (Figure 7) (Hoffman et al 1981, Su et al 2003). For entrained flow slagging gasifiers, ash softening and ash-melting or fusion temperatures are important variables. ST is related to ash particle deposition. If the temperature of the

ash particles is higher than their ST, they become sticky and tightly bond to surfaces (Miller 2005). Operating temperature exceeding FT is essential to ensure that the ash flows continuously (Hurst et al 1996). A flow temperature under a reducing environment of  $<1400^{\circ}\text{C}$  is optimal and up to  $1500^{\circ}\text{C}$  is acceptable (Patterson and Hurst 2000). Under these conditions, little or no flux is required. However, coals with the flow temperatures  $>1500^{\circ}\text{C}$ , require additional flux.



**Figure 7. Ash fusion temperature under reducing and oxidizing conditions of selected coals (Hoffman et al 1981).**

For the AFT, iron must be converted to its ferric state and all combustible matter must be burned in an ash preparation step. This is not the case in a practical gasifier under a reducing atmosphere. In the gasifier there is unreacted carbon in the ash. In addition, conditions in the gasifier are complex and impossible to simulate using a small-scale laboratory test. Therefore, the temperatures from the test should be considered qualitative (Speight 2005).

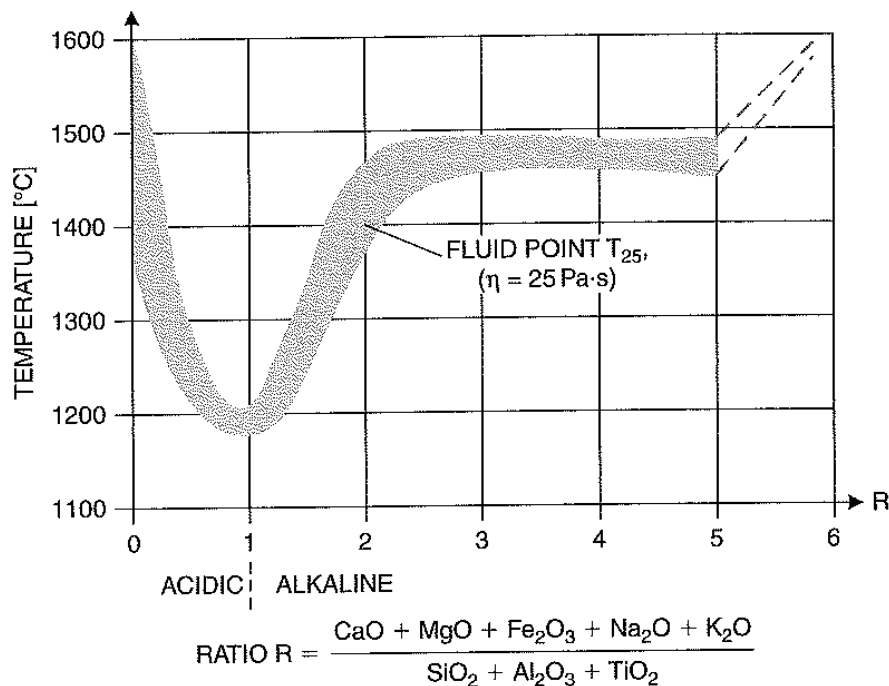
The effects of ash chemical composition on fusion temperatures were studied using regression analysis for 1250 ash samples of Eastern and Western coals (Reid 1981, Winegartner and Rhodes 1973). For gasification under reducing conditions, the ratio of silicon dioxide/iron oxide is best correlated with the temperatures, and iron oxide content is second. Under oxidizing conditions, the ratio of silicon dioxide and calcium oxide is most strongly correlated with the temperatures. Song et al (2010) investigates calcium oxide, ferric oxide, and magnesium oxide on fusion temperatures under argon using the thermodynamic computer package FactSage. The fusion temperatures of coal ash decrease with increasing calcium oxide, ferric oxide, and magnesium oxide contents then increase after reaching a minimum. As silicon dioxide/aluminum oxide ratios increase, the fusion temperatures also increase (Song et al 2010). In general, if both iron and calcium are high in coal, the softening and melting temperatures will be reduced (Higman and van der Burgt 2008).

### 3.4. Slag Viscosity

In addition to ash softening and melting temperatures, slag viscosity is an important property of ash. For slagging gasifiers, the relationship between slag viscosity and temperature is critical since it determines the flow characteristics of the slag. Coals with the same ash fusion temperature may have different slag viscosities and, therefore, behave differently in the slag gasifier (Trapp et al 2004, Higman and van der Burgt 2008).

For entrained flow gasifiers, viscosity up to 15 Pa·s is optimal, and 25 Pa·s is the maximum to keep reliable continuous slag tapping (Paterson and Hurst 2000). The temperature is represented as  $T_{25}$  because the viscosity is 25 Pa·s (Higman and van der Burgt 2008). Viscosity is strongly dependent on the temperature and chemical compositions of ash. Figure 7 shows a simplified relationship between ash fluid properties and chemical composition (Higman and van der Burgt 2008).  $T_{25}$  temperature varies with the base-acid ratio  $R$  ( $\text{CaO} + \text{MgO} + \text{Fe}_2\text{O}_3 + \text{Na}_2\text{O} + \text{K}_2\text{O} / (\text{SiO}_2 + \text{Al}_2\text{O}_3 + \text{TiO}_2)$ ).

Slag viscosity could be measured actually using a high-temperature rotational viscometer/rheometer, or it could be estimated from the composition of the ash in the gasifier feed stream. These laboratory tests are suitable for comparing coal ash slag, but it should be remembered that the measured viscosities can only be used as an indication of slag behavior in the gasifier. Other factors such as particulate carbon content, extent of iron reduction, and operating parameters may affect actual slag behavior (Patterson and Hurst 2000). The prediction method is limited because some partitioning of the ash species occurs within the gasifier, with more volatile species exiting with the gas phase (EPRI 2006).



**Figure 7. Fluid temperature of slag as a function of base-acid ratio (Higman and van der Burgt 2008).**

### 3.5. Temperature of Critical Viscosity

Viscosity is important in establishing the rate of slag flow, but the  $T_{cv}$  is equally significant in determining whether the slag will continue to flow as it cools upon leaving the slagging gasifier. As the liquid slag cools, the viscosity will initially increase linearly with the logarithm of the temperature. At the  $T_{cv}$ , the relationship of viscosity to temperature changes, and the viscosity rises more steeply (Figure 6, coal #2). Some components begin crystallization, and the slag enters the plastic intermediate zone between the liquid and solid phases (Higman and van der Burgt 2008). Some (friendly) slag has a relatively gentle initial deviation from the linear relationship, but other (unfriendly) slag has a rapid rise in viscosity at temperatures below the  $T_{cv}$ . The  $T_{cv}$  is used to set the minimum temperature for slag tapping to avoid slag tapping problems. For a slagging gasifier operating at a reasonable temperature, it is necessary that the  $T_{cv}$  be less than 1400 °C, which is preferred to allow lower gasification temperature and, thus, higher cold gas efficiencies (Patterson and Hurst 2000, Higman and van der Burgt 2008).

Predicting the  $T_{cv}$  is difficult because it depends on the separation of a crystallization phase in the molten slag. The  $T_{cv}$  may be undetected in a routine viscosity measurement until so much solid has accumulated that the slag freezes. Ilyushechkin and Kinaev (2007) studied  $T_{cv}$  with the amount of the precipitated solids using a syngas slag  $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaO-FeO}$  system and concluded that there were no direct relationships between phase compositions of melting and  $T_{cv}$ . Predicting  $T_{cv}$  based on ash chemical composition was not reliable and less useful (Reid 1981). The correlation between  $T_{cv}$  and the ash ST was poor, too. It can be improved by adding 110 °C (200 °F) to the ST (Reid 1980). The  $T_{cv}$  predicted in this method can only be used as reference.

## 4. IMPACT OF OPERATING CONDITIONS ON FLOW BEHAVIORS OF COAL SLAG IN ENTRAINED FLOW GASIFIERS

### 4.1. Slag Viscosity with Temperature and Slag Compositions

In the entrained flow gasifier, slag deposited on the wall is multi-layered and changes from fully liquid slag, partly crystallized slag, and into solid slag as temperatures decrease (Figure 5) (Jak et al 2004). Oh et al (1995) investigated the effect of temperature and the formation of crystalline phases on slag viscosity under gasification conditions. Four coal samples were used: SUFCo (Hiawatha seam, high volatile C bituminous rand), Pittsburgh #8 bituminous, and two Powell Mountain (PM) coals (unwashed PM coal-PMA and washed PM coal-PMB). SUFCo and PMA were gasification slag samples. Pittsburgh #8 and PMB were treated at 750 °C under air. Figure 8 shows how the viscosities of the four slags changed with temperature (Oh et al 1995). The viscosity of SUFCo and PMB slags gradually increased as temperature decreased, which indicates glassy slag phase (Newtonian). The viscosity of Pittsburgh #8 and PMA, on the other hand, increased rapidly as the temperature decreased, which are typical of crystalline slags (non-Newtonian). The SUFCo slag had high concentrations of silicon dioxide and calcium oxide and low concentrations of aluminum oxide and ferric oxide compared to the Pittsburgh #8 slag. Both PMA and PMB slag compositions are close to the Pittsburgh #8 slag.

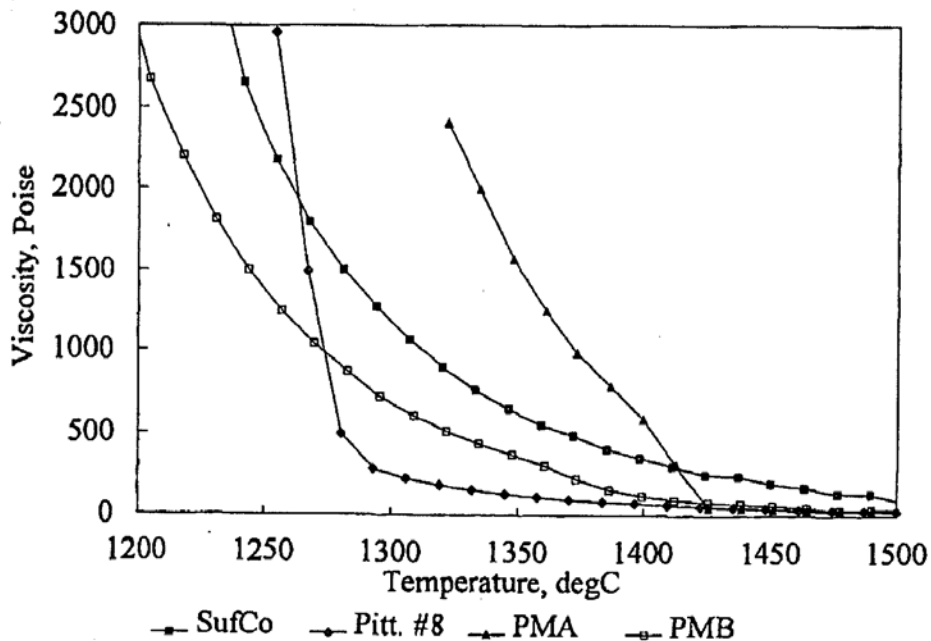


Figure 8. Slag viscosity changes with temperature (Oh et al 1995).

Slag composition strongly influences viscosity, but its mechanism is not well understood. Network theory divides individual element oxides of ash into three groups according to their effect on the silica network. The groups are network formers (increasing the slag viscosity), modifiers (decreasing the slag viscosity), and no defined roles (Kinaev et al 2007).  $\text{SiO}_2$  is the dominant network former. The alkali oxides (such

as  $\text{Na}_2\text{O}$ ,  $\text{K}_2\text{O}$ , and  $\text{Li}_2\text{O}$ ) are network modifiers. The alkaline earth oxides (such as  $\text{MgO}$  and  $\text{CaO}$ ) are usually network modifiers, but their roles are also dependant on the overall composition of the slag. The viscosity of slag with high concentrations of Ca and Mg changes sharply with temperature and sometimes results in increasing  $T_{\text{cv}}$ . Kato and Minowa (1969) used synthesized slags of  $\text{CaO-SiO}_2\text{-Al}_2\text{O}_3$  to study viscosities of the slag at elevated temperature and the effects of adding oxide, fluorides, and chlorides on the slag viscosities. The viscosity increased with increasing  $\text{Al}_2\text{O}_3$  or  $\text{SiO}_2$  contents, while  $\text{CaO}$  lowered these values. The addition of  $\text{FeO}$ ,  $\text{MnO}_2$ , or  $\text{MgO}$  lowered the viscosity of the slag. Hurst et al (1999 and 2000) measured the viscosity and  $T_{\text{cv}}$  of synthetic melts containing 5, 10, and 15 wt %  $\text{FeO}$  of the  $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaO-FeO}$  (SACF) quaternary system. Inaba and Kimura (2004) created synthetic slag from carbon-bearing  $\text{FeO}$  pellets to study slag viscosities with temperature and chemical compositions in iron and steel making. Jak et al (2004) predicted phase equilibrium and viscosity in complex coal ash slag systems based on the slag of  $\text{Al}_2\text{O}_3\text{-CaO-FeO-SiO}_2$ .

Slag viscosity with temperature and composition can be measured or predicted using empirical models based on slag composition. For entrained flow gasifiers, it is important that the models describe the viscosity of fully liquid slag in multi-component slag systems and also predict the viscosity of partly crystallized liquid slag. For predicting the viscosity of homogeneous liquid slag, the models of Urbain, Fereday, and the silica percentage model are used, as well as modified versions of those models (such as Kalmanovitch-Urbain, Watt and Fereday, and  $S^2$ ) are developed (Oh et al 1994, Browning et al 2003, and Song et al 2011). The main challenge of all these models is their validity over a limited composition and temperature range and their accuracy for different slag systems. Urbain is the most common model for predicting viscosity. The model was developed by fitting experimental data from  $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaO-MgO}$  systems to a Weymann type equation (Browning et al 2003). The equation is  $\eta = a \exp(1000b/T)$ ; where  $\eta$  is the viscosity in poise,  $T$  is the temperature in K,  $a$  and  $b$  are constants depending on the composition of the slag. For  $\text{SiO}_2\text{-Al}_2\text{O}_3\text{-CaO-FeO}$  quaternary slag systems, Urbain is the most accurate model, but it performs poorly for  $\text{SiO}_2\text{-FeO}$  binary and  $\text{SiO}_2\text{-CaO-FeO}$  ternary slag systems (Browning et al 2003). Watt and Fereday and  $S^2$  are comparable. Watt and Fereday is accurate for slags with high silica content  $>80\%$  or high iron oxide content  $>15\%$ . The  $S^2$  model is better for slags with low silica content  $<55\%$  and iron content  $<5\%$  (Browning et al 2003). In addition, the Andrade's equation is also used to express viscosity with temperature. The equation is  $\eta = A \exp(E/RT)$ ; where  $\eta$  is the viscosity in poise,  $A$  is the frequency factor,  $E$  is activation energy for viscous flow, and  $T$  is the temperature in K (Kato and Minowa 1969).

To predict the viscosity of crystalline phase slags, Annen's and Einstein's models were used (Oh et al 1995, Song et al 2011). Annen's model describes the viscosity of a mixture of the melt and crystalline phases as a function of liquid's viscosity and the solid content:  $\eta_{\text{mixture}} = \eta_{\text{liq}} (1 + 2.5c + 9.15c^2)$ ; where  $c$  is the volume fraction of solids. The viscosity of the liquid phase ( $\mu_{\text{liq}}$ ) is predicted using the Fereday model. The solid concentration and liquid composition are calculated based on thermodynamic phase equilibrium analysis. However, Annen's model is limited because it is only appropriate for slag containing very small concentrations of crystalline particles (Oh et al 1995). In addition, phase compositions in the slag can be predicted using thermodynamic equilibrium modeling tools such as the computer thermodynamic package FactSage and

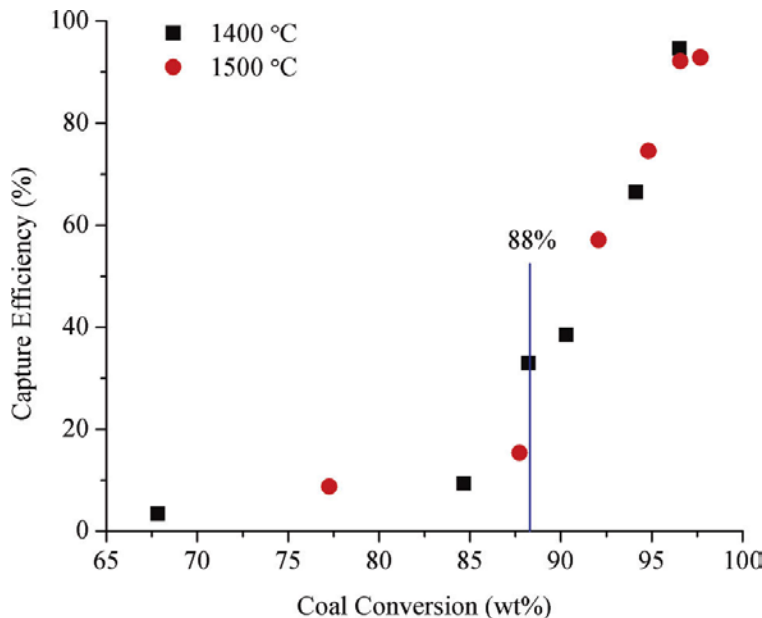
slag thermodynamic databases (Jak et al 2004, Kondratiev et al 2006, Ilyushechkin and Kinaev 2007).

Viscosity models can be used to evaluate coal suitability for entrained flow gasification. Using thermodynamic models and viscosity models together, it is possible to simulate slag flow behavior in the gasifier as a function of gas composition and operating temperature. In addition, it can be used to analyze coal flux and blends (Kondratiev et al 2006).

## 4.2. Slag Behavior with Oxygen/Carbon Ratio and Unreacted Carbon

Oxygen/carbon ratio is an important operating condition along with steam/carbon ratio, carrier gas/solid ratio, and gasification pressure (Dai et al 2008). Dai et al (2008) studied all four conditions using the pilot-scale coal entrained flow gasification system. The oxygen/carbon ratio affected the gasification temperature, carbon conversion, and syngas composition (e.g.,  $\text{CO} + \text{H}_2$  content) (Koyama et al 1996, Dai et al 2008, Cortés et al 2009). The steam/carbon ratio and carrier gas/solid ratio should be adjusted simultaneously with oxygen/carbon ratio changes (Dai et al 2008). Wu et al (2007) reported that carbon contents were 60% in coarse slag and 30–35% in fine slag because the oxygen/carbon ratio decreased in summer when ASU has a lower oxygen production.

In the entrained flow gasifier, carbon in the coal particles is converted to syngas, and mineral matters in the coal are transformed to ash/slag. The intermediate coal particles consist of unconverted carbon, minerals, and ash/slag as char-slag particles. The particles either stick (trap) on the wall or rebound (elastic reflection) from the wall, depending on the particle surface stickiness and impaction surface stickiness (Li et al 2010). Li et al (2010) studied ash deposition behavior at various conversions of a bituminous coal under gasification conditions using a laminar entrained-flow reactor and



**Figure 9. Particle collection efficiency as a function of coal conversion (Li et al 2010).**

a deposition probe. The results showed that the stickiness of pulverized bituminous coal ash is a function of carbon conversion at temperatures above the ash fluid temperature. At critical carbon conversion, large amounts of included minerals are exposed on the particle surface and melt, thus increasing the particle stickiness dramatically. The particle capture efficiency measures the ash deposition propensity and is a function of coal conversion. For Illinois #6 coal, the critical carbon conversions are about 88% at 1400 °C and 93% at 1500 °C (Figure 9). These results were similar with the coal char-slag transition under oxidizing conditions (Li and Whitty 2009). Therefore, carbon conversion is an important factor to maintain slag flow in the gasifier and limit the amount of fly ash that may cause fouling in the syngas cooler.

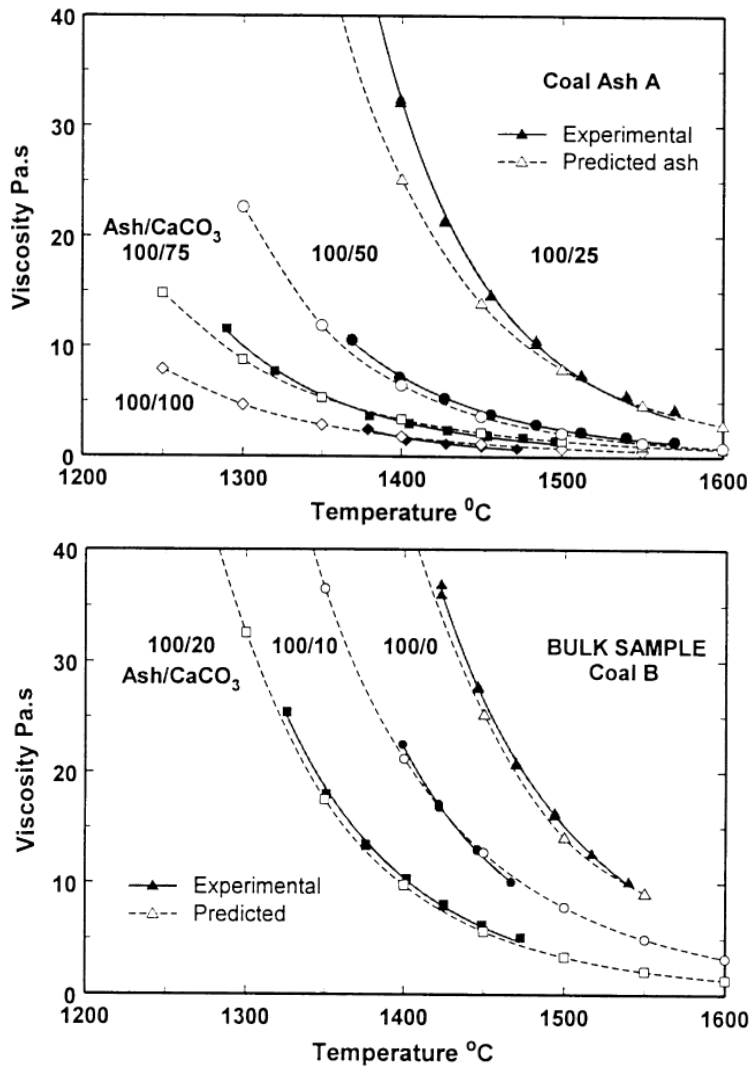
### **4.3. Viscosity with Flux Agents in Gas Environments**

In slagging operations, it may be desirable to add reagents such as fluxes to improve the flow characteristics of the slag or to adjust its chemical behavior. Possible flux agents include limestone and dolomite due to their availability, low cost, and elevated calcium and magnesium contents, which generally reduce viscosity (Duchesne et al 2010). According to the economic limit, the ash content for slurry feeding gasifiers is generally about 20% (consider evaporating water by heat) and about 40% for dry feeding gasifiers (Higman and van der Burgt 2008).

At ash flow temperatures higher than 1500 °C, a flux addition such as limestone is required (Patterson and Hurst 2000). To reduce flux cost, coal with < 3% flux requirement by weight may be blended with other coal possessing low ash fusion characteristics, thus reducing or eliminating the need for flux (Patterson and Hurst 2000). The measurements of slag viscosity versus temperature at selected levels of flux addition for Australian coal A and B are shown in Figure 10 (Patterson and Hurst 2000). The viscosity contour as a function of composition or viscosity models could be used to calculate the amount of flux required to obtain a proper viscosity for smooth slag flow in an entrained flow gasifier.

Gasification takes place in a reducing environment, with hydrogen and carbon monoxide as the dominant gases. The viscosity of Powder River Basin slag in three atmospheres was studied (Hurley et al 1996). The results showed that slag viscosity is not substantially different in air or air + 10% water vapor, but it is substantially reduced in H<sub>2</sub>/CO/CO<sub>2</sub>. Huffman et al (1980) investigated the behavior of coal ash in reducing (60% CO/40% CO<sub>2</sub>) and oxidizing (air) atmospheres using scanning electron microscopy (SEM) and XRD. The ash partially melts at temperatures 200–400 °C lower than the IDT from the AFT. Under reducing atmospheres, ash melting was faster than under oxidizing atmospheres. The percentage of melting ash increased rapidly at the range of 900–1100 °C. Ash melting is primarily controlled with iron as the flux agent. Under oxidizing conditions, potassium is the main flux agent at low temperature.





**Figure 10. Viscosity versus temperature curves for selected coal ash at various flux addition ratios (Patterson and Hurst 2000).**

## 5. REMARKS AND COMMENTS

IGCC is a potentially promising clean technology with low emissions and cost-effective carbon dioxide capture to enable power plants to continue using coal as electricity fuel (Maurstad 2005, Falcke et al 2011). For IGCC technology to become more competitive and fully commercial, low availability and high capital costs must be overcome (Maurstad 2005, Trapp 2004). In the gasification process, fouling and plugging in the syngas cooler system is the major cause of unplanned downtime (Higman et al 2006, EPRI 2006, TEC 2002). Maximizing the ash to slag ratio and minimizing the quantity of fly ash are preferred approaches to minimizing fouling problems. In addition, hot raw syngas may be cooled to convert any entrained molten fly slag to hardened solid fly ash prior to entering the syngas cooler. To improve gasification availability through an improved design and operation of the gasification process, it is necessary to understand slag behavior and the characteristics of the slagging process by both experiments and computer modeling.

Entrained flow gasifiers with high temperature have been selected for the majority of IGCC project applications. The major advantage of high temperature in entrained flow gasifiers is to generate syngas without oils and tars and remove ash as slag (Maurstad 2005). To avoid a slag blockage and keep all entrained flow gasifiers operating smoothly, slag must be steadily removed through a slag tapping device. This depends strongly on the flow behavior of the slag.

The viscosity of the slag is used to characterize slag flow behaviors and is the dominating factor that determines the probability that the ash particles will stick. Slag viscosity strongly depends on the temperature and chemical composition of the slag. Coal has varying ash content and composition, and therefore requires different operation conditions to maintain slag flow and limit downstream problems.

Coal ash content, ash fusion temperature, slag viscosity, and  $T_{cv}$  impact gasifier operating conditions and performance and are key criteria to assess a coal's suitability for an entrained flow gasifier. Due to economic considerations, the ash content for slurry feeding gasifiers is generally about 20% (consider evaporating water by heat) and for dry feeding gasifiers is about 40%. Slag viscosity of 15–25 Pa·s is required to ensure continuous slag flow. Lower  $T_{cv}$  and ash flow temperature under a reducing environment is preferred to allow lower gasification temperature and thus higher cold gas efficiency. For coals with flow temperatures higher than 1500 °C, flux must be added.

When selecting the gasifier operating temperature, consideration must be given to the ash slagging temperature or ash flow temperature of the coal, slag viscosity presented using  $T_{25}$  temperature (the temperature at which the slag viscosity is 25 Pa·s),  $T_{cv}$ , and carbon conversion. High unconverted carbon in the ash affects ash deposition on the wall of the gasifier (Li et al 2010). Fly ash with high amounts of unconverted carbon causes difficulties in downstream processing (EPRI 2006). Low temperature and high slag viscosity may solidify the slag and cause slag blockage (EPRI 2006). All considerations can be achieved by sending more oxygen to the gasifier to burn more coal and generate more heat. However, this reduces the overall efficiency of an IGCC system by increasing the parasitic power load of the air separation unit, reducing syngas yield, and shortening the life of the refractory in cases where a refractory-lined gasifier is used. For most coals, the ash melting point and slag viscosity are more constraining than carbon conversion considerations (EPRI 2006).

Slag viscosity is the most important parameter in selecting the operating temperature of the gasifier. It is strongly dependant on slag temperature and composition and can be measured or predicted using empirical models based on slag composition. For entrained flow gasifiers, it is important that the models can describe the viscosity of fully liquid slag in multi-component slag systems and also predict the viscosity of partly crystallized liquid slag. Most viscosity models have been developed based on particular viscosity datasets and, therefore, can predict slag viscosity only for the specific compositions and temperatures. Developing accurate, reliable, and general viscosity models that can be used for the multiple compositions in the slag and wide temperature ranges are necessary.

One option to reduce the slag viscosity is to raise the gasifier operating temperature. The other option is to add a flux or blend coals with low fusibility, since viscosity is affected by chemical compositions of ash. However, to raise operating temperature significantly increases oxygen demand and overall cost. Further study is needed to balance these two factors and reach high process efficiency and low costs.

Properties of coal ash and slag and operation conditions both influence slag flow behaviors. Correlating the properties and operating conditions to the behaviors of slagging could assist the selection of coals and the operating conditions for entrained flow gasifiers. Optimal operation conditions could improve both gasification efficiency and availability.

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