INCREASING REACTIVITY OF ILLINOIS COAL FOR USE IN IGCC PROCESSES

Deepak Tandon[1], Anthony A. Lizzio[1], Godfried M.K. Abotsi[2] and Gautam Saha[2]
[1]Illinois State Geological Survey, 615 East Peabody Dr., Champaign, IL 61820
[2]Department of Chemistry, Clark Atlanta University, 223 James P. Brawley Dr., SW, Atlanta, GA 30314

Introduction

The use of more reactive coals in Integrated Gasification Combined Cycle (IGCC) processes should lead to more efficient production of electricity [1,2]. In most IGCC processes (e.g., Texaco, Shell), coal is gasified in oxygen at relatively high temperatures (> 1400°C), so the intrinsic reactivity of the coal is not a critical issue. The recent development of IGCC technology that utilizes a two stage process to gasify coal (e.g., Destec process [3]), however, provides incentive for optimizing the reactivity of the coal that is added to the second stage of the gasifier (typically 20-30% of the total feed coal). Proven methods for improving the gasification reactivity of coal include adding a catalyst, e.g., calcium, to the coal [4]. Preoxidation of coal may also increase its gasification reactivity [5]. Tar formation, which is detrimental to IGCC process performance, could be suppressed by preoxidation as well as by the addition of calcium to the coal. The objective of this study [6] has been to evaluate the gasification behavior of Illinois coal for possible use in two-stage IGCC processes. This commercial application precludes the use of alkali metal catalysts such as sodium or potassium since these may volatilize and damage the gas turbines. In this paper, we determine the gasification reactivity of several Illinois coals and explore low cost methods to increase their gasification reactivity through coal preoxidation and/or addition of a suitable catalyst, e.g., calcium or iron.

Experimental

Coal and Char Preparation

Coals were obtained from the Illinois Basin Coal Sample Program (IBCSP) [7] and from selected Illinois coal mines. An Indiana coal currently being used in a commercial two-stage IGCC process was also studied. Coal chars were prepared from these coals in a 2 in. ID horizontal tube furnace (N_2, 900°C, 30°C/min, 0.5 h). After pyrolysis, agglomerated chars were ground with a mortar and pestle to a -100 mesh particle size. Some coals were preoxidized in air at 225°C for 1 h prior to gasification tests.

Catalyst Addition

Calcium (acetate) and iron (chloride) were added to the coal by ion-exchange (IE) at different pHs. IE was conducted by shaking 250 ml of each metal solution with 1 g of coal for 24 h, after which the samples were filtered and the solids submitted for metal analysis. Ca and Fe were added by IE at their natural pH (5.5 and 2.0, respectively) or at pH=10 by addition of NaOH solution.

Char Reactivity

The gasification reactivities of coal chars in 1 atm CO_2 at 850-940°C were determined by isothermal thermogravimetric analysis (TGA).

Results And Discussion

Figure 1 presents reactivity profiles (rate versus conversion) for several Illinois and the Indiana coal gasified in 1 atm CO_2 at 850°C. The reactivities of IBC-103, IBC-105, IBC-106 and IBC-108 were comparable over the entire conversion range, while IBC-101, IBC-107 and IBC-109 were most reactive. The high ash IBC-104 char and Indiana coal were the least reactive. Two Illinois coals (Industry mine and Cedar Creek) having more than twice the sulfur content (high sulfur content of the coal is important since sulfur can be recovered as a saleable byproduct), but less ash than the Indiana coal, were more than twice as reactive as the Indiana coal char. The char made from Crown II coal was nearly four times more reactive than the Indiana coal char. Transient kinetics [8,9] is being used to explain these pronounced differences in the gasification reactivity [6].

Effect of Preoxidation

Perhaps the most cost effective way to increase the reactivity of bituminous coal would be to add oxygen to the coal prior to gasification either by natural weathering or by a low temperature oxidation treatment. The reduced caking behavior and higher surface area induced by coal preoxidation has been shown in some cases to enhance the reactivity of the resultant char [5]. Figure 2 presents reactivity profiles for preoxidized and as-received coals. Preoxidation of IBC-102 and Cedar Creek coal results in little or no improvement in the char reactivity. Preoxidation of the Indiana coal actually leads to a decrease in reactivity. These results were rather unexpected. Further work is needed to optimize preoxidation conditions.

Effect of Catalysts

Figure 3 presents reactivity profiles (conversion versus time) for Ca- and Fe- catalyzed IBC-101 chars gasified in 1 atm CO_2 at 850°C. When Ca is added by IE at pH=10, there is a three fold increase in reactivity. Iron added by IE at pH=2.2 has little impact on reactivity. However, Fe added by IE at a pH=10 increases reactivity by more than a factor of two. This suggests the importance of using an alkaline medium for preparing Ca- and Fe- catalyzed coals by IE.

Figure 4 presents reactivity profiles for Crown II coal chars gasified in 1 atm CO_2 at 850°C. The as-received Crown II coal was the most reactive Illinois coal (Figure 1). When Ca and Fe are added to this coal by IE (at pH=10), char reactivity increases by more than a factor of two. The reactivities of these coals prepared at their natural pH are significantly lower, consistent with the results obtained for the catalyzed IBC-101 coal (Figure 3).

Figure 5 presents the reactivity profiles for the Indiana coal char. Addition of Ca and Fe at pH=10 results in only a slight increase in char reactivity. Addition of Ca at pH=5.5 actually decreases reactivity.

Conclusions

Illinois coals were found to be up to four times more reactive than the Indiana coal currently being used in a commercial two stage gasifier. Calcium and iron further increased the reactivity of Illinois coal, especially when IE at pH=10. Coal preoxidation was not an effective means to increase char reactivity. Preoxidation of Illinois coal prior to catalyst addition by IE would provide additional ion exchange sites for Ca and Fe and should increase both catalyst loading and dispersion.

Acknowledgments

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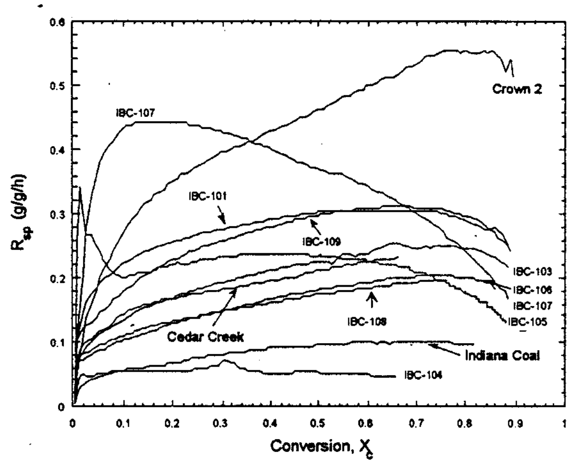

Figure 1. Reactivity profiles for Illinois and Indiana coal chars gasified in 1 atm CO2 at 850°C.

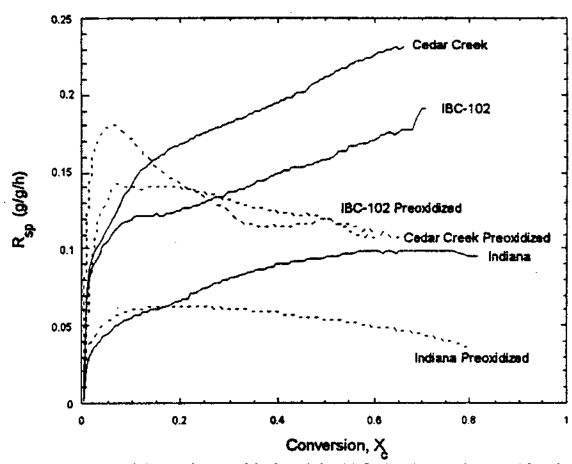

Figure 2. Effect of preoxidation (air, 225°C, 1h) on the gasification reactivity of chars prepared from two Illinois coals and the Indiana coal.

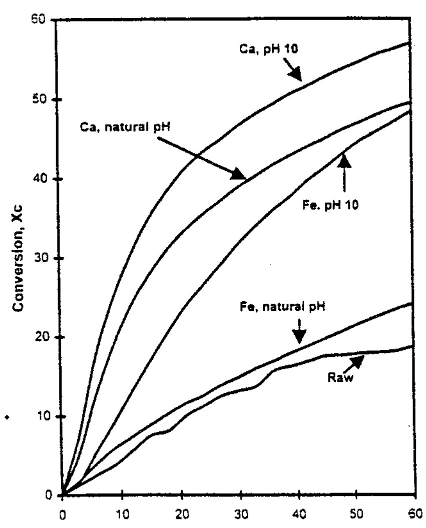

Figure 3: Effect of pH and Catalyst on Gasification Reactivity of IBC-101 Char

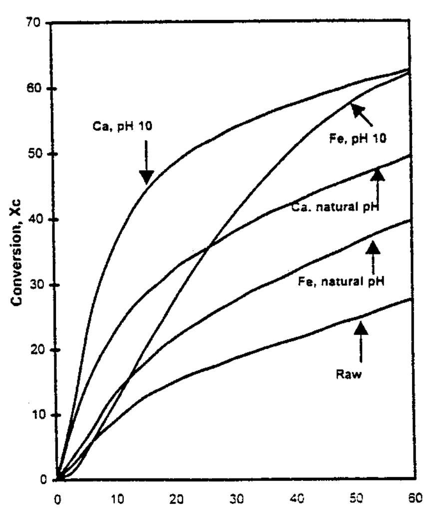

Figure 4: Effect of pH and Catalyst on Gasification Reactivity of Crown II Char

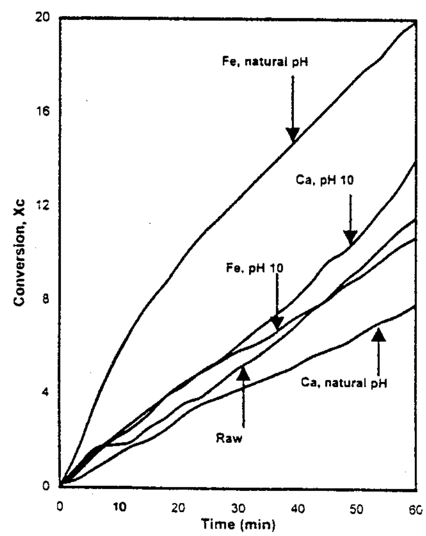

Figure 5: Effect of pH and Catalysts on Gasification of Indiana Char